Diamond Based Detectors for High Temperature, High Radiation Environments

Alex Metcalfe,[a]* George R Fern,[a] Peter R Hobson,[b,] David R Smith[b,], Gwenaelle Lefeuvre[c], Richard Saenger[d]

[a] *Wolfson Centre for Materials Processing, Brunel University London*
Uxbridge UB8 3PH, United Kingdom
[b] *Centre for Sensors & Instrumentation, College of Engineering, Design and Physical Sciences, Brunel University London*
Uxbridge UB8 3PH, United Kingdom
[c] *Micron Semiconductor Limited*
Lancing BN15 8SJ, United Kingdom
[d] *Schlumberger Limited*
91240 Clamart, France

**E-mail*: Alex.Metcalfe@Brunel.ac.uk

ABSTRACT: Single crystal CVD diamond has many desirable properties as a radiation detector; exceptional radiation hardness and physical hardness, chemical inertness, low Z (close to human tissue, good for dosimetry and transmission mode applications), wide bandgap (high temperature operation with low noise and solar blind), an intrinsic pathway to fast neutron detection through the $^{12}C(n,\alpha)^{9}Be$ reaction. This combination of radiation hardness, temperature tolerance and ability to detect mixed radiation types with a single sensor makes diamond particularly attractive as a detector material for harsh environments such as nuclear power station monitoring (fission and fusion) and oil well logging. Effective exploitation of these properties requires the development of a metallisation scheme to give contacts that remain stable over extended periods at elevated temperatures (up to 250 °C in this instance). Due to the cost of the primary detector material, computational modelling is essential to best utilise the available processing methods for optimising sensor response through geometry and conversion media configurations and to fully interpret experimental data. Monte Carlo simulations of our diamond based sensor have been developed, using MCNP6 and FLUKA2011, assessing the sensor performance in terms of spectral response and overall efficiency as a function of the detector and converter geometry. Sensors with varying metallisation schemes for high temperature operation have been fabricated at Brunel University London and by Micron Semiconductor Limited. These sensors have been tested under a varied set of conditions including irradiation with fast neutrons and alpha particles at high temperatures. The presented study indicates that viable metallisation schemes for high temperature contacts have been successfully developed and the modelling results, supported by preliminary experimental data from partners, indicate that the simulations provide a reasonable representation of detector response.

KEYWORDS: Diamond Detectors; Radiation-hard detectors; Spectrometers

Contents

1. Introduction

The broad project aim is to develop spectroscopic radiation sensors for applications where high temperature operation and radiation hardness are required, with a particular interest in detection of neutrons and gamma rays. Recent work on this has focussed on the development of diamond detectors with suitable metallisation that will survive the required operating temperatures (200-250 °C) without degradation. Previously [1] we have modelled the detector response, with a particular interest in the use of conversion media to improve thermal neutron detection, with complex geometries and have also tested diamond based sensors up to 100 °C where only a minimal change in response was observed. Here we have produced diamond sensors based on a similar metallisation scheme, packaged to allow higher temperature operation and tested them up to our design goal of 250 °C. These sensors and our alpha source have been modelled using the fully integrated radiation transport packages FLUKA2011 [2, 3] and MCNP6 [4].

2. Experimental Setup

In this following section, the diamond sensors tested, as produced by Brunel University London and Micron Semiconductor Limited are described along with an overview of computational modelling and experimental measurements.

2.1 Diamond based radiation sensors

2.1.1 Brunel sensors

A set of sensors has been produced using vacuum metallisation facilities available at Brunel University London. Commercially available 2×2×0.5 mm^3 single crystal diamond substrates marketed as "electronic grade"[1] were used. The applied contact layer was, as in previously manufactured devices, selected with high temperature stability in mind. The diamonds were mounted upon purpose designed carrier boards with a high temperature silver epoxy[2], with the front electrode connected by four aluminium bond wires. The carrier board itself is a ceramic suitable for high temperature applications[3] and has many vias included in its design to transfer heat from the copper heating block to the diamond sensor under testing. The sensor is connected through to readout by a length of 50 Ω coaxial cable rated for operation up to 250 °C.

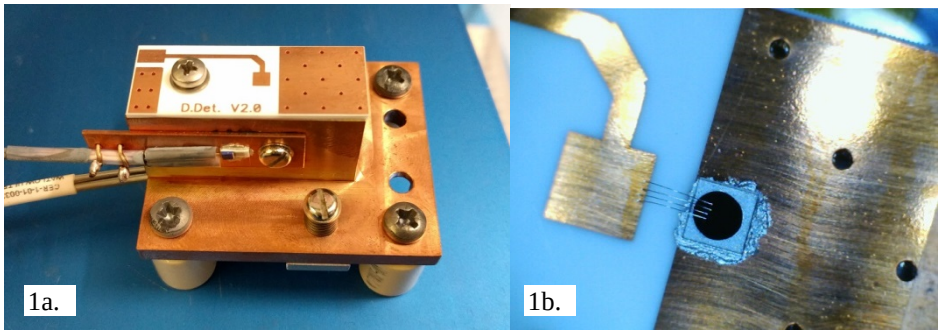

Figure 1a. A high temperature test board, mounted to the copper heating block.
1b. Magnified image of diamond attached with epoxy to a carrier board, with wire bonds.

2.1.2 Commercial sensors

A pair of prototype sensors were also supplied by Micron Semiconductor Limited for high temperature test. These devices used 4.5×4.5×0.5 mm^3 single crystal diamond substrates and were supplied with metallisations intended for high temperature operation. These sensors were tested in the packages as supplied and for connection to the amplifier a standard 200 °C rated 50 Ω coaxial cable to the feedthrough was used. This connected to the package with standard SMA connector hardware not specially rated for high temperature operation.

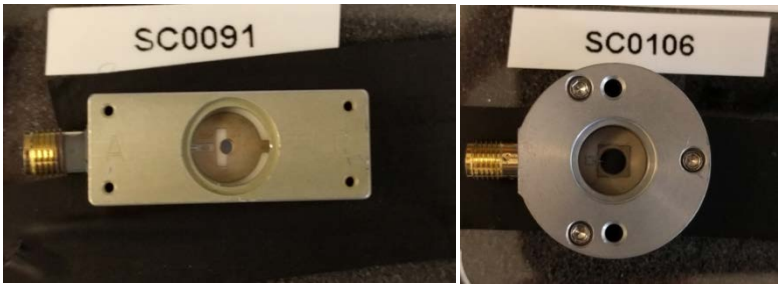

Figure 2. Commercially supplied sensors housed in their packages.

[1] As supplied by Element 6 Ltd.
[2] Duralco 120 from Cotronics Corp.
[3] 1mm thick Rubalit 708S from CeramTec GmbH.

2.2 Irradiation test setup

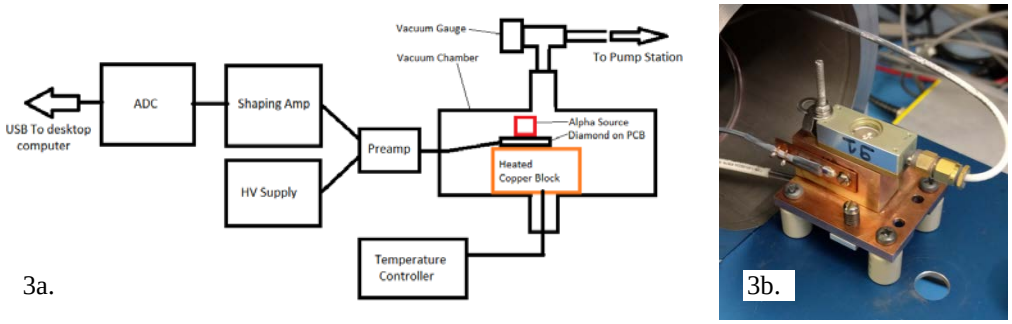

Figure 3a. Schematic diagram of equipment for alpha irradiation tests.
3b. Photograph of packaged sensor mounted to heater block.

Tests were carried out in small vacuum chamber (shown schematically in Figure 3a) evacuated by a turbopump equipped dry pumping station (to pressure < 5×10^{-4} mbar). Heating of the sensors, each mounted to a test PCB or contained within a metal package, was accomplished using a copper block with an attached AlN heater element and platinum resistance thermometer within the chamber. This was attached by multi-way vacuum feedthrough to an external temperature controller (Lakeshore 331). Electronic readout of the system was carried out by connecting internally with coaxial cable to vacuum feed-through connectors. Outside the vacuum these connect to a standard spectroscopy readout chain; first to a standard preamp (Canberra model 2004, typically used for silicon devices) then NIM crate mounted shaping amplifier (Canberra model 2021), finally connected to a PC based multichannel analyser (Canberra Eagle Plus). To assess alpha particle spectroscopy performance the sensors were exposed to a triple alpha source comprising ^{239}Pu, ^{241}Am and ^{244}Cm at approximately 1 kBq of activity each.

2.3 Monte Carlo modelling

Due to the relatively low activity of the triple alpha source available it was required to place the source close to the device under test to achieve reasonable statistics in the limited time available. This meant that there were geometrically induced limitations on the measured detector resolution. To understand this effect a model of the source and sensor has been constructed in FLUKA2011. A homogeneous block of diamond was simulated, with perfect collection in the area between the circular metal pads. The source was simulated as a cylindrical area, emitting alpha particles isotropically, filled with a compound material based on the specific activity of the isotopes used, sized with radius as directly observed from the source and thickness calculated from activity. Additionally any unique package features were simulated, including an approximation of bond wires in front of the detector.

3. Results

3.1 Brunel B-SC-6

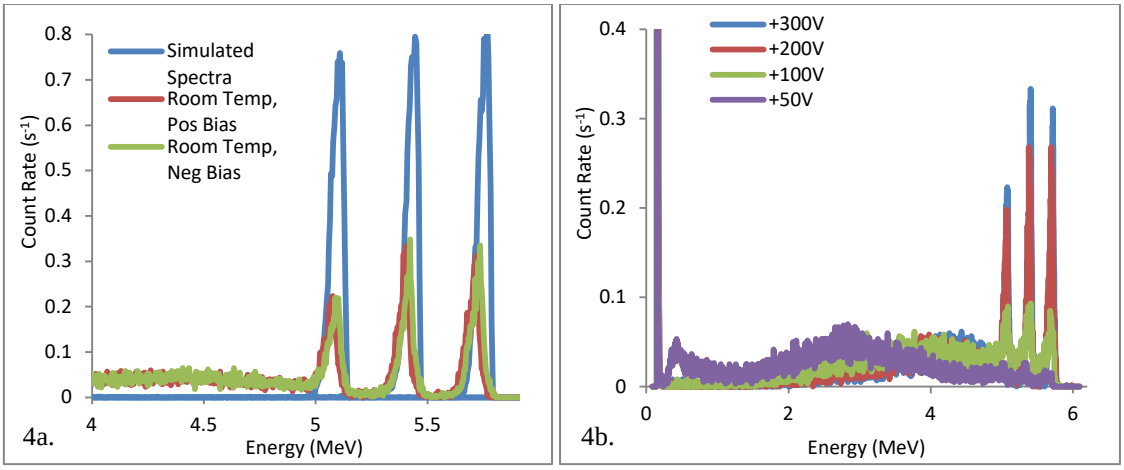

Figure 4a. A comparison of the measured triple alpha spectra, at ±300 V with model at room temperature.

4b. Measured spectra with variation of applied bias at room temperature.

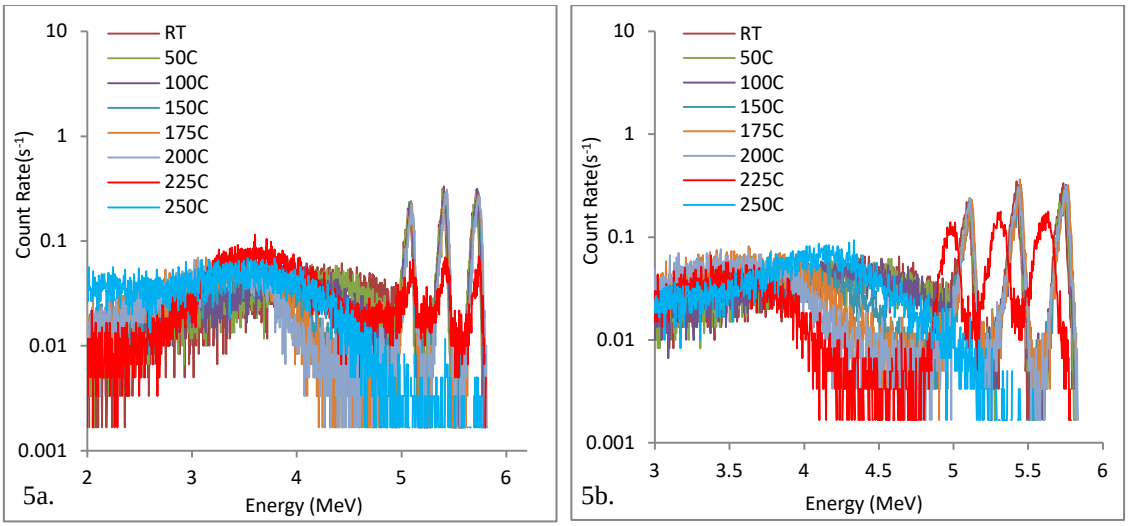

Figure 5a. Spectra with varied temperature under +300 V bias.

5b. Spectra with varied temperature under -300 V bias.

3.2 Micron SC0091

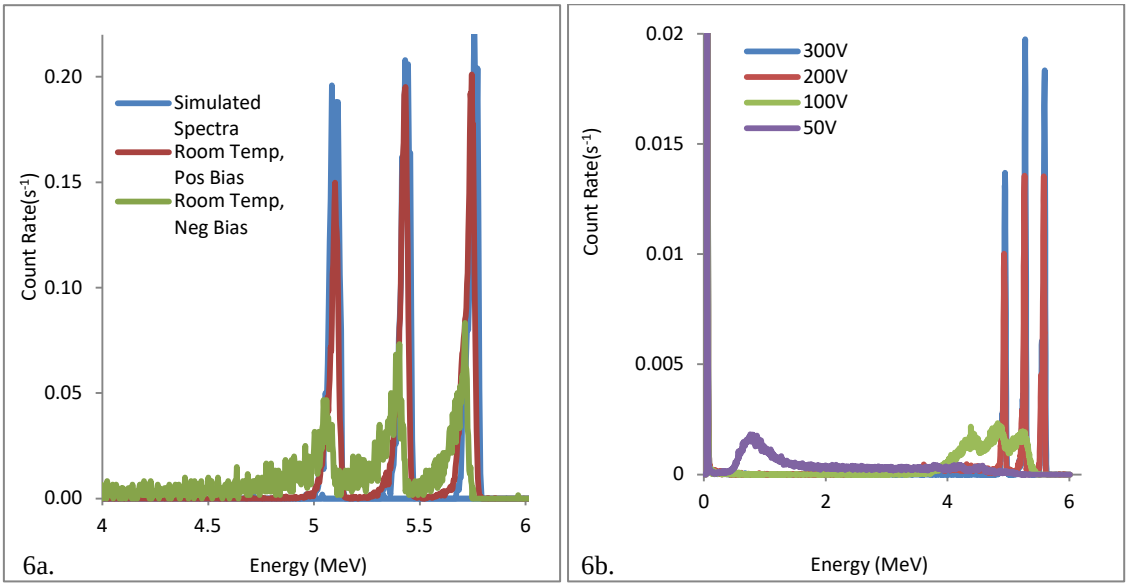

Figure 6a. A comparison of the measured triple alpha spectra, at ±300 V with model at room temperature.

6b. Measured Spectra with variation of applied bias at room temperature.

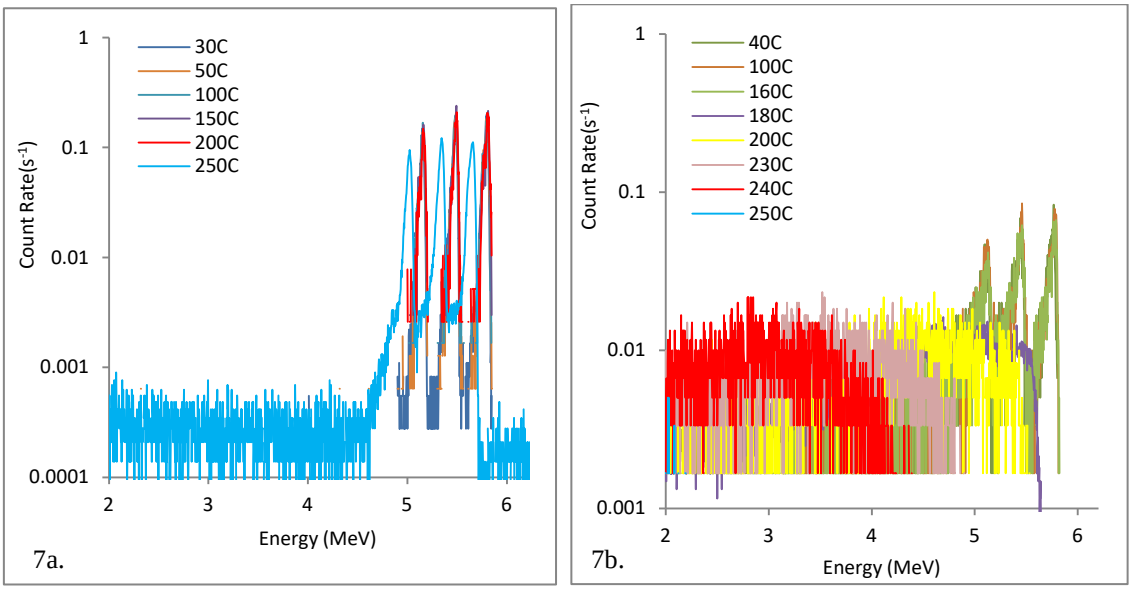

Figure 7a. Spectra with varied temperature under +300 V bias.

7b. Spectra with varied temperature under -300 V bias.

3.3 Micron SC0106

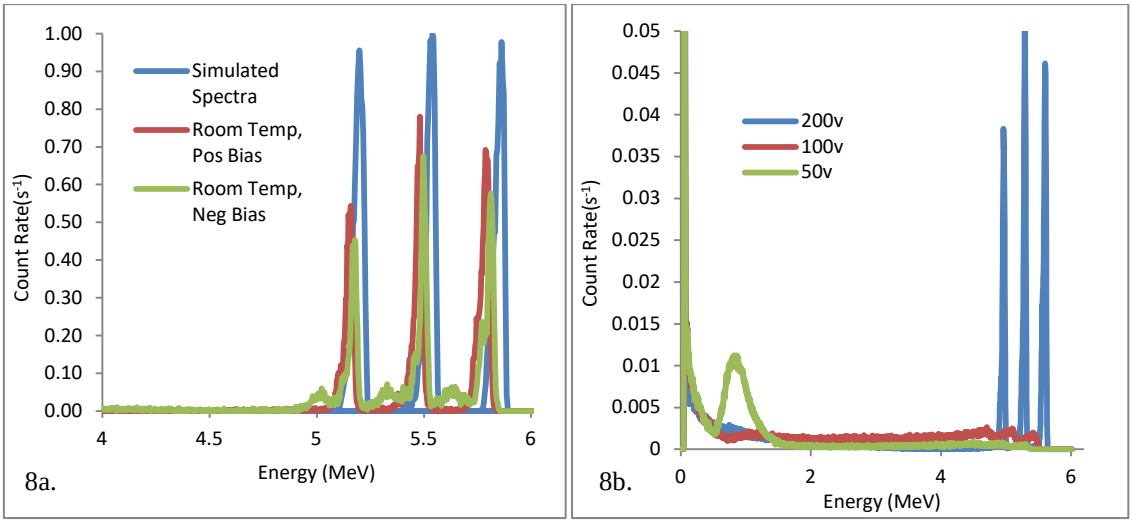

Figure 8a. A comparison of the measured triple alpha spectra, at ±200 V with model at room temperature.

8b. Measured spectra with variation of applied bias at room temperature.

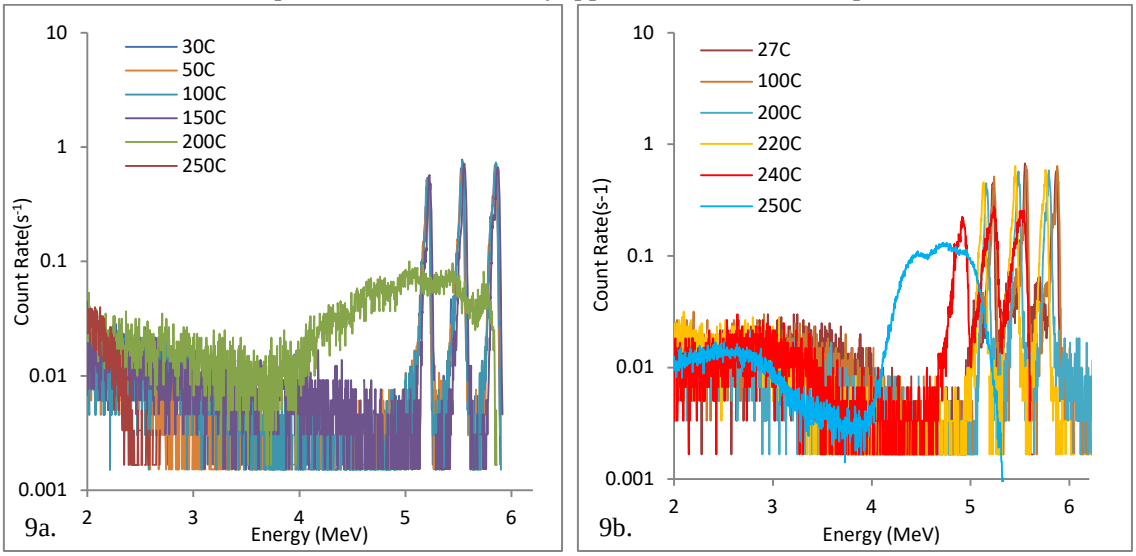

Figure 9a. Spectra with varied temperature under +200 V bias.

9b. Spectra with varied temperature under -200 V bias.

4. Discussion

Figures 4 and 5 show the results of testing diamond sensor B-SC-6 under triple alpha source irradiation from room temperature up to 250 °C. The bias is applied to the face of the sensor positioned closest to the source, hence due to the short range of alpha particles in solid matter, positive bias is hole-transport dominated and negative bias is correspondingly electron dominated. Under irradiation in both bias polarities the sensor was found to be stable under applied bias in excess of ±300 V and so, for consistency with previous work, a bias of magnitude 300 V was used. Figure 4a compares the experimental spectra to an idealised Monte Carlo model of the source and detector. This shows the experimental detector resolution to be close to the limit imposed by the geometry. The count rate from the simulated geometry

is notably higher than is observed; this is principally attributed to deviations in the setup from that simulated, due to the small source detector-distance (2.5 mm) small deviations due to alignment will lead to an appreciable difference, with the simulation representing the closest possible position and therefore the maximum count rate. This alignment issue expected to be exacerbated by the assumption of the size of the isotope deposits on the source which are simulated as 7mm in diameter. This is relatively large compared with the active area of the sensor- itself ignoring edge effects. This is repeated for the commercial detectors, shown in Figures 6a and 8a, with the same reasoning suggested, though where the package better permits consistent alignment with the source holder there is a reduced inconsistency. Up to 100 °C the detector response was unperturbed. In electron transport mode the detector response remained nearly constant up to 200 °C, with a small decrease in charge collection at 225 °C and finally a degraded signal, with no distinguishable alpha peaks, at 250 °C. With positive bias applied a similar behavior is observed, here the performance is constant up to 200 °C, after which at 225 °C a significant noise band is seen to emerge in the collected spectra, which progressively dominates over the peaks with further increasing temperature, the peaks being still readily resolvable, but becoming subsumed by the noise at 250 °C. It is suspected that this degradation at temperature is due to the interlinked effects of decreased carrier mobility and trapping-detrapping effects. It is well known that charge carrier mobility has a definite temperature dependence [5,6,7]. The incidence of this decrease in performance is also seen to correspond broadly to data from glow curves, where diamonds have been used in thermo-luminescent applications [8]

The results from the commercially produced detectors are presented in Figures 6 to 9. It was found that, at room temperature SC0106 was operable up to 200 V bias, with a large amount of noise across the whole spectra seen as the bias voltage was increased further. SC0091 was found to be stable up to above 300 V bias, which was the selected bias voltage for the main set of experimental data runs for consistency with previous work. When considering these results it is important to note that the bias is applied to opposite sides of samples SC0091 and SC0106 – facing away and towards the sample respectively. Hence for SC0091 positive bias entails transport of electrons through the bulk for collection and for negative bias holes form the majority of the signal, with the opposite being the case for SC0106.

When operated in hole sensitive mode as shown in Figures 7b and 9a both detectors can be seen to begin to severely degrade at around 180 °C, recovery from which is subject to a significant delay, contrasting with the recovery in electron sensitive mode which tracks closely enough to temperature such that any delay may be reasonably attributed to thermal lag in the system. This 180 °C degradation also broadly corresponds to thermally stimulated current and glow curve data from the wider literature [9,10]. Additionally differences in activation energies for electron and hole traps [11] would potentially explain the difference in recovery time, due to some potential re-priming effect being required for recovery of hole signal

When operated in electron transport mode (Figures 7a and 9b) both sensors tolerate higher temperatures without degradation. For SC0106 (Figure 9b) in contrast to a sudden degradation at 180 °C there is instead a gradual reduction in charge collection, evidenced by a shift in peaks to lower energies, with a notable degradation in resolution towards unresolved peaks between 230 °C and 250 °C. SC0091 (Figure 7a) can be noted to have an almost non-existent noise background until 250 °C is reached, at which point there is a small shift towards lower energy collection and a slight reduction in energy resolution. Due to the difference in detector packages, it is impossible to completely disregard that there may be some difference in the temperatures reached by the diamond substrates. However, the similar temperatures at which failure occurs in hole-transport dominated signal mode would suggest that this is not a major factor. This lack of symmetry in performance at increased temperatures is supported by work that has shown a more rapid decrease in hole mobility over electron mobility over 400 K [12]. As these mobilities have been suggested elsewhere to be highly dependent upon material bulk impurities, it may be suggested that the differences observed in performance may be due to inconsistencies in impurities in commercially available diamond substrates as much as the factors such as processing.

5. Conclusions

Diamond based radiation sensors intended for high temperature operation have been produced and tested. Of those provided by Micron Semiconductor Limited, it was possible to operate, dependent on choice of bias voltage, for alpha particles, up to 180-200 °C without significant degradation in charge collection or resolution, with operation up to 250 °C possible with some reduction in performance. This operation was obtained without use of specialised connectors and cabling specified for higher temperature operation as has been used elsewhere in the literature [13]. Future work will include further validation of high temperature coatings and operation to confirm long term stability, further work to identify causes of high temperature signal degradation, such as high temperature I-V measurements, and the development of detectors with patterned surfaces, to investigate further possible enhancements.

Acknowledgments

For funding the academic and industrial efforts (respectively) on this project the authors acknowledge the EPSRC (Engineering and Physical Sciences Research Council) (Grant no: EP/L504671/1) and InnovateUK (Grant no: HTRaD 101427). The authors would also like to thank Maria Khaleeq of the Department of Physics, Imperial College London for the wire bonding.

References

[1] A. Metcalfe, G. R. Fern, P. R. Hobson, T. Ireland, A. Salimian, J. Silver, D. R. Smith, G. Lefeuvre, R. Saenger, Development of high temperature,radiation hard detectors based on diamond, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment. (2016) http://dx.doi.org/10.1016/j.nima.2016.06.091

[2] T. Bohlen, F. Cerutti, M. Chin, A. Fosso, A. Ferrari, P. Ortega, A. Mairani, P. Sala, G. Smirnov, V. Vlachoudisl, The FLUKA code: Developments and challenges for high energy and medical applications, NUCLEAR DATA SHEETS 120 (2014) 211–214.

[3] A. Ferrari, P. Sala, A. Fasso, J. Ranft, FLUKA: a multi-particle transport code cern-2005-10, CERN200510 (2005), INFN/TC 0511, SLACR773.

[4] T. Goorley, M. James, T. Booth, F. Brown, J. Bull, L. J. Cox, J. Durkee, J. Elson, M. Fensin, R. A. Forster, J. Hendricks, H. G. Hughes, R. Johns, B. Kiedrowski, R. Martz, S. Mashnik, G. McKinney, D. Pelowitz, R. Prael, J. Sweezy, L. Waters, T. Wilcox, T. Zukaitis, Initial MCNP6 release overview, Nuclear Technology 180 (3) (2012) 298–315.

[5] J. Isberg, A. Lindblom, A. Tajani, D. Twitchen, Temperature dependence of hole drift mobility in high-purity single-crystal cvd diamond, Physica status solidi (a) 202 (11) (2005) 2194–2198.

[6] H. Jansen, D. Dobos, V. Eremin, H. Pernegger, N. Wermes, Temperature dependence of charge carrier properties in single crystal cvd diamond detectors, Physics Procedia 37 (2012) 2005–2014.

[7] J. Pernot, C. Tavares, E. Gheeraert, E. Bustarret, M. Katagiri, S. Koizumi, Hall electron mobility in diamond, Applied physics letters 89 (12) (2006) 2111.

[8] E. Vittone, C. Manfredotti, F. Fizzotti, A. L. Giudice, P. Polesello, V. Ralchenko, Thermoluminescence in cvd diamond films: application to radiation dosimetry, Diamond and related materials 8 (7) (1999) 1234–1239.

[9] D. Briand, P. Iacconi, M. Benabdesselam, D. Lapraz, P. May, C. Rego, Thermally stimulated properties of cvd diamond films, Diamond and Related Materials 9 (3) (2000) 1245–1248.

[10] E. Pace, A. De Sio, L. Tozzetti, M. Bruzzi, Diamond dosimeters: applications to radiobiology and space environments, Rendiconti Lincei 25 (1) (2014) 87–90.

[11] M. Marinelli, E. Milani, A. Paoletti, G. Pucella, A. Tucciarone, G. V. Rinati, M. Angelone, M. Pillon, Analysis of traps in high quality cvd diamond films through the temperature dependence of carrier dynamics, Diamond and related materials 12 (10) (2003) 1733–1737.

[12] C. J. Wort, R. S. Balmer, Diamond as an electronic material, Materials Today 11 (1) (2008) 22–28.

[13] R. Pilotti, M. Angelone, S. Loreti, G. Pagano, M. Pillon, F. Sarto, M. Marinelli, E. Milani, G. Prestopino, C. Verona, G. Verona-Rinati, High temperature performances of cvd single crystal diamond detectors, Proceedings of Science 2015.