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Diamond-based radiation detectors for very high dose rate environments– 16207

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ABSTRACT

Diamond detectors offer a novel technology allowing the non-destructive assay of highly radioactive environments in the civil nuclear sector. Four detectors of different manufacturing origins have been characterised using dark current measurements and dose rate response in controlled environments. Results show the electrode material and contacts to the electrode are critical in achieving a reliable and stable detection device. This work demonstrates that diamond detectors offer considerable promise for civil nuclear applications, where high dose rate measurements are required.

INTRODUCTION

Background

Legacy facilities previously used for the reprocessing of spent nuclear fuel at Sellafield Ltd (UK) represent significant financial liabilities for the taxpayer. One of the challenges faced is the corrosion of ageing and contaminated pipework and vessels. Therefore, in preparation for the upcoming Post-Operational Clean Out (POCO) phase, accurate radiological characterisation of these facilities is required to inform a cost effective and safe decommissioning strategy [1]. Due to the highly active nature of some of the facilities, physical access is not desirable as it may reduce containment of the waste and extend the duration of the POCO phase, resulting in additional cost. Therefore, characterisation will only be possible with innovative technologies able to perform Non-Destructive Assay (NDA) of the facility through existing access points. Typically the only access points currently available for remote inspections are of small diameter penetrations in containment walls.

Such access restrictions are not the only challenge in highly active environments; the dose rates anticipated within these facilities are extremely high with most electronic devices unlikely to survive for meaningful durations. Conventional radiation detectors including silicon, gallium arsenide (GaAs), high purity germanium (HPGe) or cadmium zinc telluride (CZT) devices become saturated and damaged to the point of failure [2]. Therefore, innovative and novel technologies are required; i) to measure the radiation dose rates present in these facilities, and ii) to accurately identify the radionuclides present. This paper presents background

and results on high quality chemical vapour deposited (CVD) diamond as a detector material for measuring dose rates in highly active environments, with future work aimed at expanding the technology in developing a spectroscopic system.

Diamond as a radiation detector

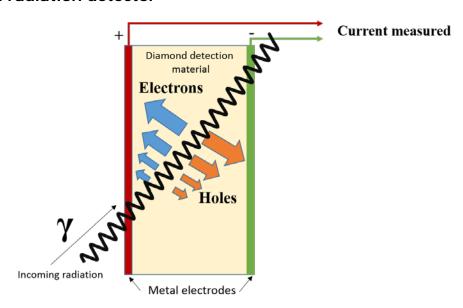


Figure 1. Principle of radiation detection in a diamond detector

Diamond behaves as a radiation detector in a similar manner to many other semiconductor based devices [3]. When ionising radiation enters the diamond detector, electrons are promoted from deep within the valence band to the conduction band, with this charge drifting under an electric field to biased metallic electrodes on either side of the diamond wafer [3], as shown in Figure 1. This charge can be measured, and its magnitude is indicative of the energy deposited in the detection material by the incoming ionising radiation. The charge collected at the electrodes for a single incident photon is too small to be measured directly unless amplified: a typical alpha particle from Am-241 has an energy of 5.486 MeV, which, if completely collected, generates roughly 422000 electron-hole pairs.

Without amplification, this would be impossible to measure directly; however should many particles of ionising radiation arrive continuously then this signal becomes a significant measurable current, known as leakage current. Dose is a measure of the energy deposited in a material which manifests as mobile charge carriers [4]; therefore the current generated as a result of irradiation is believed to be proportional to the dose rate [5], and independent of the individual photon energy [6], or radioisotope producing the radioactive particles. This measurement of current to determine dose rate is the focus of this paper, rather than amplification and detection of single incidents (spectroscopy).

In recent years, the use of synthetic diamond detectors for medical dosimetry measurements has been considered due to their tolerance to high dose

environments and their tissue equivalence [7]. Additionally, due to its radiation tolerance, diamond has been studied as a particle detector by the RD42 collaboration at CERN [8,9]. These studies have shown that diamond offers an improvement in high dose resilience when compared to conventional radiation detectors [2].

The wide band gap (5.5 eV) of diamond means that it has a theoretical zero idle counting rate as the thermally generated signal is negligible [10]. This temperature stability permits the detectors to be very small without the need for large cooling devices required for other detectors. Such size and radiation hardness make diamond the ideal candidate for performing NDAs in the civil nuclear sector, especially at Sellafield Ltd.

This paper focuses on the characterisation and leakage current testing of four separate diamond detectors all based on single crystal CVD diamond. A description of the detectors is first provided, followed by the method of characterisation and testing. Results from characterisation and testing are provided along with a discussion of the results, including the planned progression for this technology.

DESCRIPTION

Four different detector prototypes based on single-crystal CVD diamonds (4.5 x 4.5 x 0.5 mm) were evaluated for their suitability to be used as a leakage current detector. Table I gives details of the four devices, two of the devices (S1 and M1) were commercially available devices manufactured by Diamond Detectors Limited (DDL) and two devices (F1 and T1) were manufactured at the University of Bristol. All detectors had co-axial connectors. Images of the four devices are shown in Figure 2.

TABLE I. Details of the four different diamond detector devices.

Device	Manufacturer	Electrode Material	Bonding material	Electrode Diameter	Casing Material
S1	DDL	Gold	Gold wire bond	4 mm	Brass
M1	DDL	Gold	Gold wire bond and silver conducting adhesive	4 mm	Brass
F1	University of Bristol	Aluminium	Silver conducting adhesive	4 mm	Plastic
T1	University of Bristol	Aluminium	Silver conducting adhesive	3 mm	Plastic

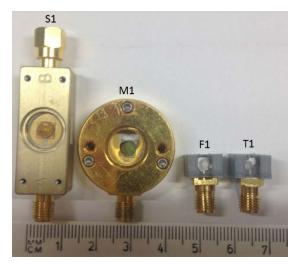


Figure 2. Images of the four diamond detector devices tested.

METHOD

Dark Current Characterisation

An initial test of performance, especially in the characterisation of the metal coating contact, is to measure the dark current [11], which is attributed to the small electric current that flows through a detector device when no incident particles are arriving at the device. These measurements were performed in an electrically shielded die-cast aluminium box. The voltage was varied in 25 V steps between -1000 V and +1000 V and then returned in 25 V steps from +1000 V to -1000V, repeated twice, giving a total of four voltage sweeps (two increasing and two decreasing). The bias voltage was changed every 30 seconds and current recorded every 0.05 ms, giving a total of 600 data points for each voltage step. The mean current was plotted with standard deviation shown as error bars, calculated from the middle 200 (10 second) period to avoid current fluctuations related to the switching of bias voltage value.

Dose Rate Measurement

Response to irradiation was performed at two facilities: i) 100kV photon beam produced by an X-ray tube at Singleton Hospital, Swansea, UK and ii) the self-shielded model 812 Co-60 irradiator at the Dalton Cumbria Facility, Cumbria, UK (DCF) which had an activity of approximately 170 TBq during experiments.

For the photon beam, the radiation dose rate was calculated using the inverse square law after raising the tube vertically away from the detector from a well characterised position. For the Co-60 irradiator a motorised stage was used that had movement in the x, y and z directions. This allowed travel of the sensor in multiple directions whilst irradiation was taking place, including a 90mm range of movement away from the source. A Radcal ion chamber was used in conjunction with the diamond sensors to determine the dose received by the diamonds during irradiation. All four diamonds were tested using the Co-60 irradiator and the x-ray

source. In all experiments the bias voltage was applied and current recorded at 0.05s intervals. The bias voltage was set to +300 V for S1 however for detectors M1, F1 and T1 this was reduced to +100 V due to instability issues, discussed in dark current characterisation results.

RESULTS AND DISCUSSION

Dark Current Characterisation

The dark current measurements for the four diamond detector devices are shown in Figure 3. These show there are significant differences between the four devices, with S1 showing a relatively uniform and linear pattern where the current does not vary by more than 0.5 nA over the whole voltage range. This is representative of a good metal electrical contact and is highly indicative of a reliable detector. The other 3 detectors are notably different. M1 appears to have the worst performance, with large variations in current as the voltage is both increased and decreased from 0 V. F1 and T1 also have large variations in current at the high and low voltages however they appear more stable in the middle range of voltages (-400 V to + 400 V). The upper current is always approximately 1 mA or –1 mA, this is a safety feature of the bias supply as it will stop applying a bias voltage if the current is

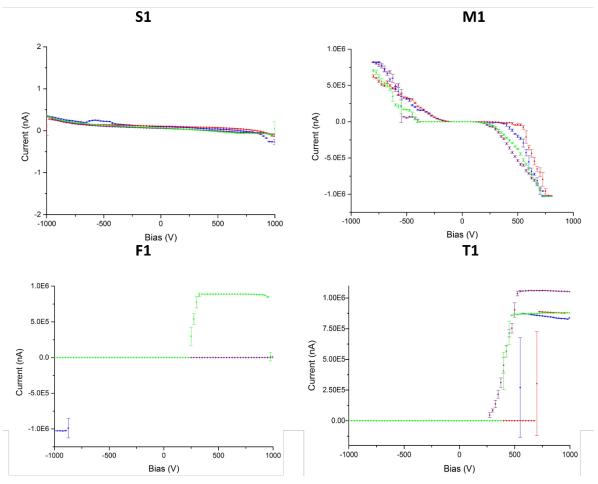


Figure 3. Dark current measurements for the four diamond detector devices.

greater than 1 mA. Therefore, once the current reaches either 1 mA or -1 mA there will be no further increase in current regardless of voltage applied.

From these observations it is clear that the manufacture of a diamond detector device is not straightforward, with the metal electrodes and contacts needing to be of high quality to avoid polarisation effects and ensure mechanical adhesion. The metal contacts on S1 and M1 are the same material, however S1 was connected using wire bonding whereas for M1, a conductive adhesive was used that appears to be unsuitable. The same conductive adhesive was also used for F1 and T1 that potentially explains the instability observed in F1 and T1.

Dose Rate Measurement

The current response of the four diamond detectors when exposed to a variety of dose rates at DCF are shown in Figure 4.

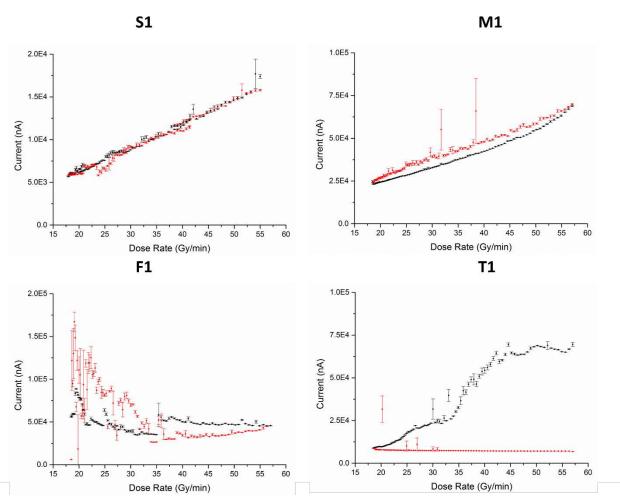


Figure 4. Current versus dose rate for the four diamond devices tested at the Dalton Cumbria Facility.

Mean current at each dose rate has been plotted for each detector, with standard deviation shown using errors bars as an indication of stability. S1 shows a linear current response related to dose rate, correlating with dark current measurements, Figure 3. F1 and T1 show poor linearity with dose rate, and the currents recorded are much greater. This is evidence of a poor detector device that is unreliable. This also correlates strongly with the dark current measurements as these showed several areas of instability and large background currents. M1 shows a similarly linear response which was unexpected as the dark current measurements suggested that this was a poor detector, however the reason for this linearity may be that the bias voltage applied was only +100 V, where the dark current measurements were stable. However, such a low bias voltage is not optimum for detector operation. These results highlight that although a detector responds in a linear fashion to dose rate, it does not necessarily make the detector suitable for use and a combination of dark current measurements and controlled dose measurements are required to determine the suitability of a detector for reliable use.

Each device was then tested using a 100 kV X-ray source giving up to 5 Gy/min in order to assess detector behaviour at lower dose rates. The general trend of increasing current with dose rate was demonstrated by all four detectors, with linearity shown by S1, M1, and T1. A possible explanation for the poor response shown by F1 is that the silver contacting adhesive is a poor electrical bond to the diamond's aluminium contacts, which supports the data obtained at DCF in Figure 4. Previous irradiation at DCF may have contributed to spalling of the silver adhesive from its diamond contact.

Although made using silver adhesive, detector T1 exhibited linear behaviour at these low dose rates, suggesting some promise in this regime. Indeed, this low dose rate operation is supported by data shown in Figure 4 in the initial (black) run at the lower dose rates.

The mode of x-ray tube emission meant that the flux of x-rays was expected to vary; this explains the significant current fluctuations recorded flowing through the diamond detectors during irradiation.

Many of the outlying data points exhibit large error bars, indicating the detector current was unstable at the time of measurement. If necessary in future measurements these data points may be discounted as unsuccessful measurements.

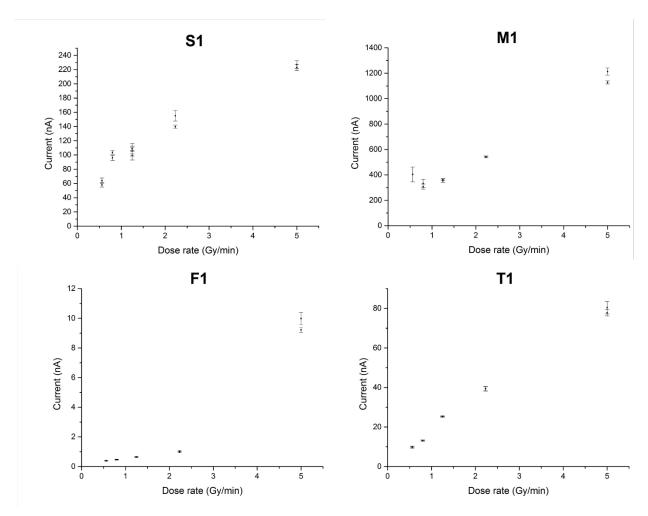


Figure 5. Current versus dose rate for the four diamond devices tested at the Singleton Hospital X-ray source.

CONCLUSION

The performance of four single crystal CVD diamond detectors of different manufacturing origins were assessed for their suitability to measure high radiation dose rates in civil nuclear applications. This was achieved by characterisation of the dark current response over a range of bias voltages to determine the quality of the metal contacts and connections, and then testing in controlled radiation environments to determine current response to a variety of dose rates.

It was found that the connections to the metal contacts were critical in the stability and reliability of the detector device, with wire bonding far more preferable to conductive adhesives. The detector with wire bonding contacts showed good stability and a linear response to increasing dose rate, which is promising for radiation monitoring purposes up to very high radiation intensities. The other three

detectors using conductive adhesive contacts showed less promise due to instabilities in dark current and associated poor performance in dose rate measurements. Detector M1 showed a linear response to dose rate, however this was due to the detector being biased at only +100~V which is far less than optimum (approximately $1~V/\mu$ m). Use of such a detector in civil nuclear applications would not be desirable due to potential reliability issues.

The combined data from Figure 4 and Figure 5 shows that for high dose rate operations, detectors S1 and M1 is more reliable than T1 and F1. The importance of this reliability cannot be understated in civil nuclear applications, and stability enhancement through electrical contact optimisation is predicted to be the key to realising this goal.

Overall, there is strong promise for development and deployment of diamond detectors in the civil nuclear sector, with one detector already proven to be stable and have a linear response to increasing dose rate up to very high gamma intensities. A method for testing the reliability and stability of diamond detectors has also been evaluated and future work will apply this method to a variety of other detector variants manufactured to determine the most reliable and repeatable method of detector manufacture.

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