CVD Diamond X-ray Detectors for Radiotherapy Dosimetry

S P Lansley[†,*], G T Betzel, J Meyer
Department of Physics & Astronomy
University of Canterbury
Christchurch, New Zealand

[†] Also, MacDiarmid Institute for Advanced Materials &
Nanotechnology, New Zealand
[*] stuart.lansley@canterbury.ac.nz

F Baluti
Oncology Service
Christchurch Hospital
Christchurch, New Zealand

L Reinisch
Department of Physical & Earth Sciences
Jacksonville State University
Jacksonville, AL, USA

Abstract— **X-ray detectors were fabricated from a range of commercially-available synthetic diamond fabricated using chemical vapour deposition (CVD). As these detectors are intended as dosimeters for use in radiotherapy (beam calibration and profiling, in-situ dose measurements etc.), they were appropriately packaged and tested in a clinical environment, using clinical apparatus and following clinical procedures. The combination of linear dose-rate dependence of the photocurrent, negligible dark current levels (pA or less, compared to nA photocurrents), low priming doses (few Gy) and high specific sensitivities (of up to 460 $nCGy^{-1}mm^{-3}$, compared to reported values of 50–140 $nCGy^{-1}mm^{-3}$ for a commercial natural diamond-based X-ray detector) demonstrates the potential of these devices as simple‒to‒use, small size, tissue-equivalent, sensitive X-ray dosimeters.**

I. INTRODUCTION

Radiation detection and dosimetry play an important role in radiation environments such as hospital x-ray imaging and treatment facilities. Dosimetry is used during system calibration to assess beam characteristics for later use in treatment planning, but could also be used during patient exposure to confirm the exposure dose [1]. For radiotherapy, an ideal dosimeter has the following features: high accuracy – the ability to indicate physical dose correctly; high precision – the reproducibility of results under similar conditions; low detection limit – the lowest dose detectable; measurement range – it should be able to detect radiation over an appropriate dose range; linear dose response – readings should be linearly proportional to the given dose; dose-rate independence – readings should be independent of the dose-rate; energy independence – readings should be independent of the radiation energy; and high spatial resolution – it should allow the measurement of the dose in a very small volume [2].

Diamond has been proposed as a material for the construction of radiation detectors for many years, for reasons including its near-tissue equivalence – its atomic number (Z = 6) is close to that of tissue (Z ≈ 7.4) – and radiation hardness. Being a solid state material with high atomic density, it should be possible to realise small-volume detectors suitable for obtaining measurements with high spatial resolution. Also, it is expected that the response of detectors fabricated from diamond should be independent of the x-ray energy and dose rate.

Early reports utilized carefully selected natural diamonds [3,4]. Natural diamond-based detectors for radiotherapy applications are commercially-available [5,6], but they are not widely used due to poor availability and high cost arising from the scarcity of suitable high-quality material. Recent developments in the synthesis of diamond have led to both chemical vapour deposition (CVD) [7-11] and high pressure high temperature (HPHT) [12-14] diamond being considered for radiotherapy dosimetry. The use of synthetic diamond should make possible the fabrication of cheaper diamond-based x-ray detectors with more reproducible characteristics, resulting from the possibility of controlling the quality of the diamond during synthesis.

II. EXPERIMENTAL METHODS

A. Material

Commercially-available free-standing synthetic diamond films were purchased from three manufacturers; unless otherwise stated, the material was synthesised using chemical vapour deposition (CVD).

Black, opaque polycrystalline films 5 × 5 mm^2 and 100, and 200 μm in thickness were obtained from Diamonex [15]. These films were 'as grown' with random crystallite

orientation and hence exhibited considerable surface roughness on the growth surface. The diamond films purchased from Diamonex, and devices fabricated using these films, will hereafter be referred to as 'Diamonex' or 'Dx' followed by the film thickness.

Transparent, colourless 5×5 mm^2 and 100, 200 and 400 µm in thickness were obtained from Diamond Materials GmbH [16]. These films had been polished and were described as being of 'optical quality'. The diamond films purchased from Diamond Materials GmbH, and devices fabricated using these films, will hereafter be referred to as 'Diamond Materials' or 'DM' followed by the film thickness.

Two nominally undoped ([N] < 1 ppm, [B] < 0.05 ppm) single-crystal films were obtained from Element Six Ltd [17]. One film had been polished on one side (average surface roughness, R_a < 10 nm) and lapped on the other side (R_a < 250 nm), whilst the other film had been polished on both sides (R_a < 30 nm) [18]. Both were transparent and colourless. These films were 3×3 mm^2 and 500 µm thick. The diamond films purchased from Element Six Ltd, and devices fabricated using these films, will hereafter be referred to as 'Element Six' or 'E6', followed by 'SC' (for single-crystal) and either 'PL' (polished/lapped) or 'P2' (polished both sides).

B. Device fabrication

Sandwich-type device structures were obtained using thermal evaporation of silver contacts (~ 200 nm thick) on each side of the diamond films. A shadow mask was used to define the circular contacts; 2-mm diameter was used for the 5×5-mm^2 films (Diamond Materials and Diamonex) and 1-mm diameter was used for the Element Six 3×3-mm^2 films. Device parameters are summarised in Table I.

TABLE I.   SUMMARY OF DEVICE PARAMETERS.

Material	Thickness (µm)	Contact (mm ∅)	Volume (mm^3)	Field[a] (V µm^{-1})
Diamonex				
Dx100	100	2	0.31	2.50
Dx200	200	2	0.63	1.25
Diamond Materials				
DM100	100	2	0.31	2.50
DM200	200	2	0.63	1.25
DM400	400	2	1.26	0.63
Element Six				
E6SCPL	500	1	0.39	0.50
E6SCP2	500	1	0.39	0.50

a. When biased with 250 V from the Farmer 2570/1 Dosemeter.

Each device was glued on to the end of a narrow strip of printed circuit board (PCB) with copper tracks running the length of it. Electrical connection was made between these tracks and the silver contacts on the diamond film using short pieces of fine wire and conductive silver epoxy. At the opposite end of the PCB, physical and electrical connections were made to a triaxial bulkhead connector. The device-PCB-bulkhead assembly was housed inside a Perspex enclosure of total length about 200 mm and internal diameter about 6 mm (Fig. 1). For most of the length of enclosure the wall thickness was about 3 mm, but at the device end it was about 1 mm. The external dimensions of the shaft of this enclosure
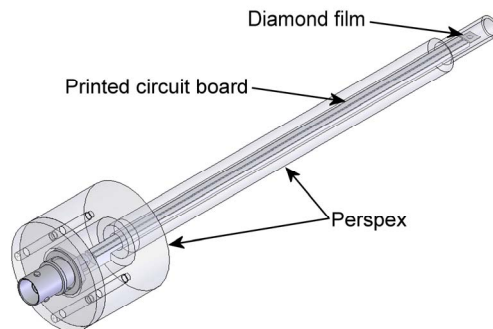

Figure 1. Schematic of device enclosure.

were chosen to match the external dimensions of a Perspex build-up cap used with thimble-type ionisation chambers at Christchurch Hospital, allowing the packaged diamond devices to be used under the same conditions as those ionization chambers. A near-tissue-equivalent paraffin-based wax [19] was used to fill the cavity around the device for all devices except DM200 and E6SCP2; this is done to eliminate air from the cavity in order to avoid partial loss of electronic equilibrium and hence minimize fluence perturbations [1].

C. Device characterization

The detectors were characterised using 6 MV photons from a Varian Clinac 600C treatment linear accelerator in the Oncology Service at Christchurch Hospital, Christchurch, New Zealand. As the detectors are being developed for radiotherapy applications, they were tested using the same irradiation geometry and parameters as are typically used for beam calibration on this linear accelerator.

The device under test was inserted into a hole at a depth of 10 cm in a solid water phantom ~$30 \times$ ~30 cm$^2 \times 18$ cm deep, providing 10 cm of build-up material between the source and the device. The solid water phantom was placed on the linear accelerator couch such that the device was positioned at the linear accelerator isocentre; 100 cm source–device distance. A 10×10 cm^2 field size was used for all measurements. The gantry angle was kept at 0°, i.e. the source being directly overhead. The detector was held in a horizontal position such that the x-rays entered the device through the electrical contact to which the bias was applied.

The physical arrangement used permitted the replacement of the diamond detector with a calibrated thimble-type ionisation chamber (0.6 cm^3 Farmer ion chamber) for assessment of the beam output. A dose of approximately 1 centigray (cGy)/monitor unit (MU) was measured using the same solid water phantom, source–device distance and field size; the permitted calibration tolerance for clinical application is ±2 percent from this value. Therefore, the linear accelerator preset nominal dose rates of 50, 100, 150, 200, and 250 monitor units per minute (MU min^{-1}) correspond to dose rates of approximately 0.5, 1.0, 1.5, 2.0, and 2.5 gray per minute (Gy min^{-1}), respectively.

Electrical measurements were performed using two different electrometers; one was the standard electrometer available at Christchurch Hospital, a Farmer 2570/1 Dosemeter, and the other a Keithley 6430 Subfemtoamp Remote SourceMeter. In both cases, the electrometer was

situated outside the treatment room and triaxial cabling through the treatment room wall provided the electrical connection between the detector and the electrometer.

The Farmer 2570/1 Dosemeter was used to bias and measure the output of the detector. It can be set to provide a bias of about 250 V or about a half, quarter, or eighth of this bias. The limited range of biases available did not permit the use of a constant electric field for our range of devices and hence only the full bias (~ 250 V) was used, resulting in different electric fields in the devices depending on the diamond film thickness (see Table 1). The dosemeter was operated manually in the 'charge' mode and the output was read out as the charge (nC) measured by the dosemeter over a user-specified time interval, i.e. a time integral of the current through the device. Therefore only an average current during that user-specified time interval could be obtained, not an accurate reading of the current in the detector at any particular instant. Within the 'charge' mode, the user can select either a 'low' or 'high' range, where these ranges have the following limitations: the 'low' range has a full scale of 20.475 nC, a resolution of 0.005 nC, and a maximum continuous input rate of 6 nA; the 'high' range has a full scale of 204.75 nC, a resolution of 0.05 nC, and a maximum continuous input rate of 60 nA. Hence the average current resolution (and detection limit) for a t-second time interval is either $5/t$ pA for the 'low' range or $50/t$ pA for the 'high' range.

The Keithley 6430 Subfemtoamp Remote SourceMeter was also used to bias the detector (it can provide any voltage between -210 and +210 V) and measure detector current over time. It was used for its high current sensitivity and its temporal resolution. The SourceMeter was controlled remotely via National Instruments LabVIEW 7.1.

III. RESULTS & DISCUSSION

Prior to irradiation, the dark current of each detector was measured using the Farmer 2570/1 Dosemeter. For the Diamond Materials and Element Six detectors, the dark current was below the detection limit of the Farmer Dosemeter (1.25 pA when a 4-second time interval and the 'low' range setting were used); this simplifies the measurement of the detector response. However, dark currents of several nanoamperes were measured for the Diamonex detectors.

Diamond detectors generally require pre-irradiation (priming) in order to stabilise the detector signal [20]. Element Six and Diamond Materials devices (excluding DM100) required only 10 Gy or less to be fully primed, whereas the Diamonex devices and DM100 were never fully primed after doses of many tens of gray; see [21] for a discussion on the priming of these devices. The devices were primed using a dose rate of 250 MU min^{-1} (~2.5 Gy min^{-1}).

A. Transient response

The transient response of the 'primed' detectors, when exposed to alternating periods with and without exposure to the x-ray beam (with increasing dose rates, as indicated by the text at the top of the figure), is shown in Fig. 2. The horizontal error bars indicate the 4-second intervals during which the charge was measured.
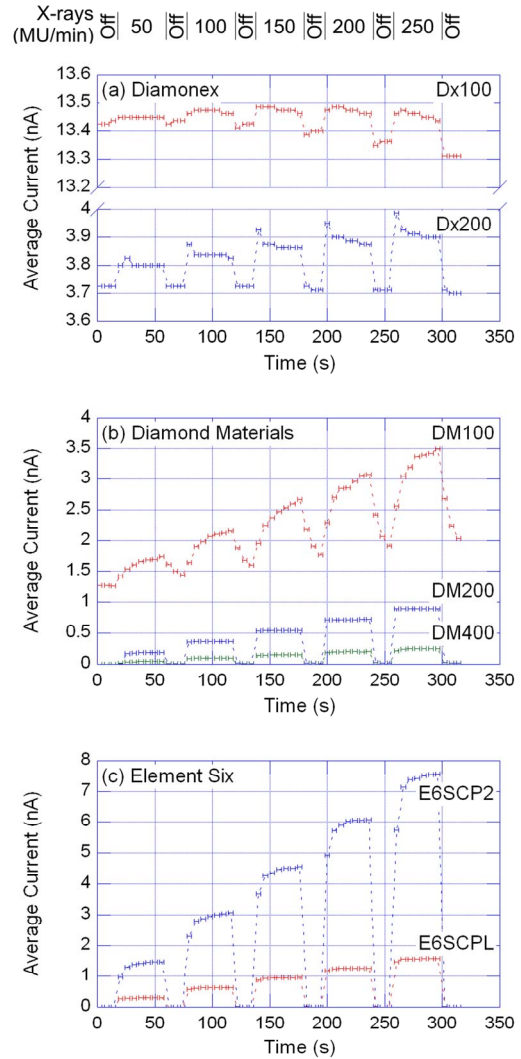

Figure 2. After-priming responses of (a) Diamonex, (b) Diamond Materials, and (c) Element Six detectors.

Diamonex devices exhibit large, changing dark currents, upon which small photocurrents are observable by the change in current when the x-ray beam is switched on and off (Fig. 2(a)). A brief overshoot was seen with device Dx200 when the x-ray beam was turned on. Diamond Materials devices DM200 and DM400 showed a step-like change in current was observed when they were irradiated with x-rays (Fig. 2(b)); 'zero' dark current was still observed. DM100, however, had slow rise and fall times, never quite reaching a steady state when the x-ray beam was on or off. Element Six devices also continued to show 'zero' dark current. Like the Diamond Materials devices, they responded in step-like manner when irradiated with the x-rays, but appeared to have slightly slower rise times than the Diamond Materials devices (Fig. 2(c)).

From the data in Fig. 2, the steady-state photocurrent (i.e. the steady state of the current after subtraction of the background dark current) was calculated as a function of dose rate for each device (Fig. 3). Photocurrent (I_{ph}) is expected to be dependent on dose rate ($\dot{D}$) according to a power-law relationship ($I_{ph} \propto \dot{D}^{\Delta}$), where the exponent ($\Delta$) usually lies
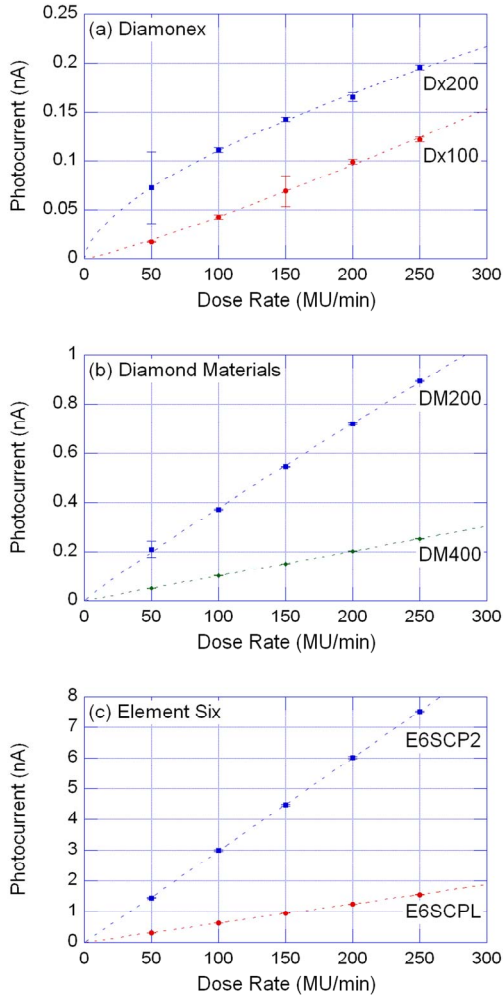
Figure 3. Steady-state photocurrent as a function of dose rate for (a) Diamonex, (b) Diamond Materials, and (c) Element Six detectors.

obtained for the Diamonex detectors lie outside this range, possibly due to the material quality and/or errors in the measurement of the photocurrent arising from low photocurrent levels and decreasing dark currents.

The sensitivities of the three detectors with 'linear' responses – DM400, E6SCPL, and E6SCP2 – were calculated to be about 6, 37, and 178 nanocoulombs per gray (nC Gy^{-1}), respectively. Linear curve fits could be used for the other devices – Dx100, Dx200, and DM200 – however these fits would not pass through the origin. Over the dose rate range reported here (50–250 MU min^{-1}), approximate linear sensitivities of about 3, 3.5, and 20 nC Gy^{-1} were measured; R^2 was greater than 0.998 for each of these three data sets.

Specific sensitivities of detectors are often reported in the literature as they take into account the size of the detector; specific sensitivity is the detector sensitivity divided by the sensitive volume of the detector. For the detectors reported here, specific sensitivities of about 10, 6, 32, 5, 94, and 454 nanocoulombs per gray per cubic millimetre (nC Gy^{-1} mm^{-3}) were obtained for detectors Dx100, Dx200, DM200, DM400, E6SCPL, and E6SCP2, respectively; the electrical contact area and the manufacturer's quoted film thickness were used to determine the sensitive volume.

TABLE II.   SUMMARY OF DEVICE PFERFORMANCE.

Material	Exponent Δ	Approx. linear sensitivity (nC Gy^{-1})	(nC Gy^{-1} mm^{-3})
Diamonex			
Dx100	1.16 ± 0.05	3.14	10.0
Dx200	0.61 ± 0.02	3.54	5.64
Diamond Materials			
DM100	N/A	N/A	N/A
DM200	0.94 ± 0.02	20.2	32.2
DM400	1.00 ± 0.01	5.94	4.72
Element Six			
E6SCPL	1.00 ± 0.01	37.0	94.3
E6SCP2	1.01 ± 0.01	178	454

between 0.5 and 1.0; Δ = 0.5 if there are no traps or when the excitation rate is so high that traps are unimportant, while Δ ≈ 1 if there are uniform or quasi-uniform trap distributions over a range of depths in the forbidden energy gap [22]. Power-law curve fits are shown through the data in Fig. 3.

The exponent values (Δ) for the curve fits shown in Fig. 3 are listed in Table II; R^2 values were greater than 0.997 for the Diamonex devices and 0.999 for the Diamond Materials and Element Six devices. Three devices – DM400, and the two Element Six devices – have exponents extremely close to unity. A unity power-law exponent is an extremely desirable characteristic of a detector, as it means that the detector response is linearly dependent on the dose rate without any offset or threshold; this significantly simplifies calculation of the dose rate from the detector photocurrent.

A review of the literature reveals a range of reported exponent values: 0.92–1 for PTW natural diamond detectors [23-28], 0.86–1.035 for CVD diamond detectors [23,24,29-31], and 0.49–0.97 for HPHT diamond detectors [12,13]. The values obtained for the Diamond Materials and Element Six devices lie close to one and compare well to the values reported for CVD diamond by other authors. The values

A wide range of specific sensitivity values have been reported in the literature for diamond detectors: 50–140 nC Gy^{-1} mm^{-3} for PTW natural diamond detectors [7,23,25,26,32]; 18–164 nC Gy^{-1} mm^{-3} for detectors fabricated from high-pressure-high-temperature (HPHT) synthesised diamond [13]; and from just a few to over a thousand nC Gy^{-1} mm^{-3} for CVD diamond-based detectors [7-9,23,24,29,30,33,34]. A wide range of sources and energies have been used to characterise diamond detectors, including ^{60}Co, ^{90}Sr, and electrons (4–25 MeV) and x-rays (250 kV, and 6, 10, and 25 MV) from linear accelerators. For comparison, Bruzzi *et al.* [35] reported the following typical values for standard on-line dosimeters: standard 0.6 cm^3 Farmer ionisation chamber, 0.036 nC Gy^{-1} mm^{-3}; miniature EXRADIN T1 ionisation chamber, 0.028 nC Gy^{-1} mm^{-3}; Scanditronix GR-p BS silicon diode, 474 nC Gy^{-1} mm^{-3}; and Scanditronix SFD stereotactic diode, 353 nC Gy^{-1} mm^{-3}. Other values reported include 330 nC Gy^{-1} mm^{-3} for an epitaxial SiC diode [35] and 128–480 nC Gy^{-1} mm^{-3} for polymer-based detectors [36].

The sensitivity, and hence specific sensitivity, will depend considerably on the device geometry, in particular the contact area and film thickness, as well as the bias or electric field applied, complicating the direct comparison of reported values. Doubling the contact area and halving the film thickness will not change the device volume but is likely to change the sensitivity, and hence specific sensitivity, of the device for various reasons, including: a reduction in the distance over which the x-rays can interact with the material; a reduction in the device resistance due to the combination of increased cross-sectional area and reduced thickness; and an increase in the electric field if a fixed bias is used. The values reported here were obtained with a range of electric field values from 0.5 to 2.5 V μm^{-1}, as listed in Table 1.

B. Fixed-dose response

The charge generated in detector E6SCP2 was measured for fixed doses in order to determine whether the detector can distinguish between dose just one or two percent different. Fig. 4 shows the mean charge measured for doses in the range 98–102 MU at a dose rate of 250 MU min^{-1}; the error bars indicate one standard deviation. It can be seen that the detector can distinguish between these relatively close doses. However, there is some overlap of data once data beyond one standard deviation are considered. A linear fit with a gradient of about 2 nC MU^{-1} is drawn through the data; this corresponds to a sensitivity of about 200 nC Gy^{-1}, comparable to the value reported above for this device.

The charge generated by a fixed dose of 100 MU at different dose rates was also measured (Fig. 5(a)); again, the error bars indicate one standard deviation. Ideally, the charge measured for a fixed dose should be independent of the dose rate, but it is obvious that this is not the case here. In order to deliver a certain dose at 50 MU min^{-1} it takes five times as long as delivering that dose at 250 MU min^{-1}. Fig. 5(b) shows the mean current through the device (i.e. the charge measured divided by the delivery time) as a function of the dose rate. A power-law fit [ref] is shown through the data; this fit yielded an exponent (Δ) of 0.93 ± 0.01 and R^2 of greater than 0.9999.

C. Cumulative dose response

Using the Keithley 6430 Subfemtoamp SourceMeter, detector E6SCP2 was biased at various voltages (100, 150, and 200 V) and the current recorded at approximately 1-second intervals as the x-ray beam (250 MU min^{-1}) was
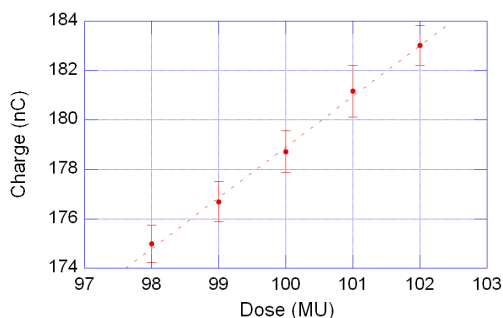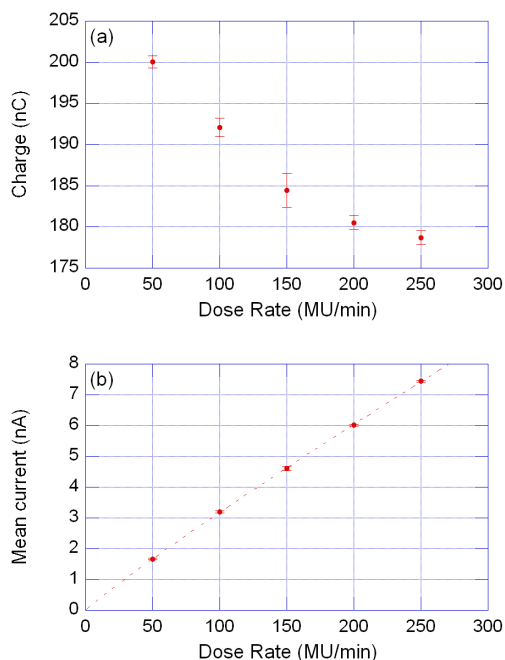
Figure 5. (a) Charge and (b) mean current measured for a dose of 100 MU at various dose rates; error bars show one standard deviation.

switched on and a dose of over 10 Gy was given to the detector. Numerical integration was then used to determine the cumulative charge measured in the detector as a function of the cumulative dose received by the detector. Ideally, there should be a linear relationship between the dose received and the charge measured. This was true once the dose received had exceeded about 1 Gy. At low doses, though, there was some non-linearity. Fig. 6 shows the charge for doses up to 2.5 Gy; only every third data point is shown for clarity.

A curve fit corresponding to an exponential current rise has been plotted through the data; R^2 values are greater than 0.9999. These fits yield steady-state current values of 4.96, 6.08, and 7.06 nA for biases of 100, 150, and 200 V, respectively. These values correspond to sensitivities of about 119, 146, and 170 nC Gy^{-1}, respectively, in the linear region (dose greater than about 1 Gy). Exponential rise constants of 0.314, 0.328, and 0.353 s^{-1} were obtained for biases of 100, 150, and 200 V, respectively, corresponding to rise times (time for signal to increase from 10 % to 90 % of the steady-state signal) of about 7.0, 6.7, and 6.2 seconds, respectively.
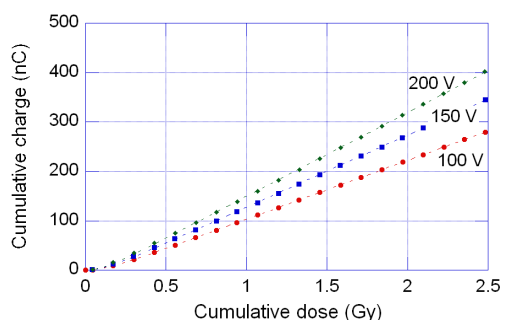
Figure 4. Charge measured for fixed doses at a dose rate of 250 MU min^{-1}; error bars show one standard deviation.

Figure 6. Cumulative charge as a function of cumulative dose; curve fits correspond to an exponential current rise.

IV. CONCLUSIONS

A range of x-ray detectors have been fabricated from commercially-available CVD diamond and characterised using 6 MV x-rays from a clinical linear accelerator.

The polycrystalline material obtained from Diamonex is not suitable for the fabrication of x-ray detectors for the following reasons: large varying dark currents (on the order of nanoamperes or greater) ; extremely high priming doses, as priming never full occurred after exposure to several tens of gray; relatively small photocurrents (sub-nanoampere), resulting in low sensitivity; and non-linear relationship between photocurrent and dose rate. These characteristics would make the accurate and consistent determination of x-ray dose extremely difficult.

The material obtained from Diamond Materials (excluding DM100) and Element Six appear far more suitable for x-ray dosimetry, due to the following desirable characteristics: extremely low, and hence effectively zero, dark currents (below 1.25 pA); low priming doses (less than 10 Gy); higher sensitivity; and linear relationship between photocurrent and dose rate.

Further characterisation of one Element Six detector (E6SCP2) showed that it is capable of distinguishing between doses one-hundredth of a gray different and has a slight dose-rate dependence for doses up to about a gray.

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