Evolution of Diamond based Microdosimetry

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Abstract. The requirements for solid state microdosimetry particularly within the context of medical and space environments necessitate a tissue equivalent and radiation hard material, for which diamond is uniquely suited. An overview of the current status of diamond based microdosimetry is given. This overview will explore previous and current technologies developed by the Centre for Medical Radiation Physics (CMRP). The overview will analyse technologies in terms of their advantages and disadvantages within the context of microdosimetry. Using this as a basis, recommendations with respect to fabrication methodologies/techniques are provided in order to direct the course and progress of development so as to achieve true diamond based microdosimetry.

1. Introduction
Diamond based radiation detector technologies is a relatively new area of study in the field of solid state microdosimetry. This interest in diamond is attributed to its radiation tolerance [1, 2] and tissue equivalence [3-7]. Diamond based microdosimeters whilst still under development, are under consideration for application in hadron therapy and radioprotection applications in harsh radiation environments where their radiation hardness will enable long term operation.

Microdosimetry is a methodology which considers stochastic radiation interactions and associated biological effects for Sensitive Volumes (SV) on the scale of the cell. Key to microdosimetry is the concept of lineal energy (y) which is defined as the quotient of an energy deposition event (s) and the mean chord length of the sensitive volume (l) [8]. Microdosimetric Spectra are typically plotted in a semi-logarithmic scale as events span orders of magnitude. The two most commonly used spectra are frequency (yf(y) vs. log(y)) and dose (y2f(y) vs. log(y)) distributions, where equal areas under the curve represent equal fractions of events and fractional doses respectively. The experimental determination of these quantities then requires detectors/microdosimeters with accurately defined micron sized SVs and thus a well-known mean chord length (l).

At the Centre for Medical Radiation Physics (CMRP), University of Wollongong, a variety of different designs and fabrication processes have been utilised in the attempt to realise an array of well-defined and accurately defined micron scaled SVs. In this paper, an overview of the current status of diamond based microdosimetry at CMRP is given. In addition, an examination of the most promising fabrication technologies currently available that might be applicable to diamond microdosimetry will be examined. This work will feature the various technologies currently available and will assess each with respect to the fabrication techniques utilised in their creation within the context of their ability to satisfy the aforementioned solid state microdosimeter requirements.
2. Fabrication techniques for diamond microdosimetry

Whilst there is extensive literature available regarding the fabrication of diamond-based radiation detectors, the area of diamond microdosimeters remains relatively untouched. This section will detail the positive and negative implications of the more promising techniques currently available.

2.1 Ion implantation

Electrically isolated SV structures may be created by means of ion implantation. This technique involves the controlled bombardment of energetic charged ions to create a buried electrode structure. The depth of structure is determined by the ion energy as defined by the Bragg peak. The first use of this technique for the purpose of microdosimetry is outlined by Davis et al. 2012 [9] in the creation of a metal-insulator-p-type diamond (M-I-P) structure. Ion implantation of 2 MeV boron through a patterned aluminium mask was utilised to create a 3D wall-less SV structure at approximately 1.38 µm depth in Type IIa electronic grade single crystal diamond from Element Six. The fabrication methodology employed in the creation of this device was successfully used to create an electrically isolated array of SVs with scalability to achieved SVs appropriate for microdosimetry, albeit with very low charge collection efficiency. This under-performance of the device with respect to CCE has been hypothesised to be the result of radiation induced damage via the boron implantation technique. The normal damage process due to ion implantation in diamond is the creation of vacancies and interstitials, which may act as charge traps and scattering sites. Annealing is generally sufficient to remove such damage and electrically activate the implanted impurities. The lower than expected CCE indicates that either the implantation or annealing methodology requires further investigation in order to be useful for diamond-based devices.

2.2 Laser ablation

Physically isolated volumes may be created via laser ablation as opposed to the electrical isolation achieved via ion implantation. This technique was utilised to create a conceptually simple planar metal-insulator-metal (MIM) structure utilising high purity single crystal chemical vapour deposited (scCVD) diamond sourced from Element6 [10]. Laser ablation is utilised to create ‘trenches’ surrounding the planar Metal-Insulator-Metals structure in order to achieve SV definition. The benefits of this technique is that SV definition may be defined post-growth, through lithographic and laser milling processes. It should be noted that whilst SV may be reduced in two dimensions, there is still a dependence upon the thickness of the sample due to planar structure of the device. The primary advantages of laser ablation over alternative techniques such as deep reactive ion etching, is the rate and ease at which diamond may be micro-machined to a desired depths and patterning. This device demonstrated good charge confinement and true promise in terms of applicability as a diamond based microdosimeter. The devices have demonstrated charge signal amplitudes matching the theoretical value, i.e., approximately 100% CCE. The IBIC experiments showed that the technique of pulsed laser milling is capable of creating physically isolated SVs, leading to reduced effect of charge sharing between adjacent pixels. Charge collection confinement was shown to be affected by particle energy and angle of incidence, i.e., penetration depth. In addition, it was shown that the charge collection confinement could be further improved/optimized through appropriate adjustment of the electric field strength. Lastly, it was shown that a combination of the fabricated laser milled trenches and surrounding electric fields when the device is in operation mode results in a well-defined and laterally confined charge collection region. However, the planar MIM structure means that there is an inherent dependence upon the sample thickness.

2.3 Active Brazing Alloy

The use of lateral electrode type structures in diamond detectors is a relatively new area of research. One of the more prominent methods has been in the utilisation of femtosecond pulsed lasers to create graphitic columns/pillars within the diamond bulk, and whilst ideal in terms of tissue equivalence, leads to many issues in charge collection properties [11]. One alternative is the uses of
commercially available Active Brazing Alloys (ABA). These materials commonly gold or silver based contain an active ingredient (1-5%), to promote reaction and wetting/adhesion with ‘ceramic’ surfaces. For applications involving diamond, ABAs containing titanium are the active ingredient of choice given their closely matched work function ($\phi=4.33$ eV) and this ability to form a carbide. ABAs are a versatile material, able to coat surfaces and/or fill voids. The use of ABAs in combination with patterned laser ablation provides a simple and effective means for creating lateral electrodes within diamond. The performance of Silver ABA for metallisation of diamond for radiation detection applications was shown by Davis et al 2016 [12]. In this paper, silver ABA was used to fill laser milled regions to create a 3D lateral electrode structure (3D-LES). Unlike the two previous prototypes, this is the first design to employ a lateral electric field structure. This device achieves 100% CCE in the region of interest and along with laser ablated boundary trenches is able to successfully isolate charge collection from surrounding regions [12]. Whilst the 3D-LES detector is not yet ready for microdosimetry, it does appear to satisfy the requirements of a radiation detector for high energy physics experiments, i.e. radiation hardness, fast detection and large charge collection distances and efficiency. With the appropriate adaptations of the designs, ABA technologies is suitable for the creation of microdosimetric technologies. Although it should be noted that applications within photon dosimetry where tissue equivalence is paramount, the use of high Z metals like silver and titanium would not be desirable.

2.4 Multi-layered diamond

The common issue with previous designs discussed, is the dependence upon the diamond sample thickness, resulting in SV structures too large to be applicable for microdosimetry. Whilst it is possible to purchase thin diamond (10-30µm), the samples are extremely fragile and liable to fracture even under the small amounts of force utilised in contact-based photolithography. To that end, CMRP is instead investing its efforts in the development of bi/tri-layered diamond structures, such as has already been developed by the University of Rome Tor Vergata laboratories [13]. The bi-layered structure requires the growth nominally intrinsic (Type IIa) diamond upon low grade commercial single crystal High Pressure High Temperature (HPHT) diamond. The tri-layered structure utilises a two-step diamond deposition process upon low grade commercial single crystal High Pressure High Temperature (HPHT) diamond. The first deposition phase involves the growth of single crystal Boron Doped Diamond (BDD) upon the HPHT substrate. The second phase is the growth of nominally intrinsic (Type IIa) diamond, upon the layer of BDD. The thickness of each layer can be customised, given a known growth rate. The end result is a layer of high quality diamond with customisable thickness.
The use of layered diamond growth processes in combination with suitable post processing (Laser ablation and/or Dry Reactive Ion Etching etc) provides the most reliable means or creating a pixelated diamond microdosimeter device. To that end, CMRP has produced a prototype diamond micro-dosimeter utilising a combination of laser ablation and active brazing alloys upon a bi-layered structure as a proof of concept for next generation diamond microdosimetry. The 3D-LES-2 structure features cylindrical SV structures created by laser ablation and ABA situated within a thin membrane of electronic grade pCVD diamond grown at the Melbourne Centre for Nanofabrication (MCN). Patterned laser ablation is used to create an array of concentric circles to form the outer electrode structure with further laser ablation used to connect rows of outer electrode in parallel. A final round of laser ablation is used to mill hollow cylinders at the centre of each of the aforementioned concentric circles, followed by the application of silver ABA to replace ablated diamond and create electrical contacts. Currently the minimum size of the inner electrode is restricted such that the diameter is greater or equal to 50µm to allow for direct wire bonding. In the case of the bi-layered device, the depth of laser ablation was performed such that both the inner and outer electrode penetrated into the low-quality diamond substrate. To better illustrate the concept of the 3D-LES-2 device, a conceptual schematic is depicted in Fig. 1. Electrical characterisation of the device has now been completed with charge collection characterisation to be performed in the near future. The results of this study will be detailed in an upcoming paper.

3. Discussion and Conclusion
Diamond is a highly interesting material for radiation detectors in a variety of applications from high energy physics to medical physics. CMRP is at the forefront of 3D pixelated detectors for the purpose of microdosimetry. Diamond based radiation detectors can be developed utilising a variety of different fabrication techniques. Previous device structures have had SV size dependent upon the thickness of the diamond sample or a loss in CCE in the case of ion implantation. The future of diamond based microdosimetry will utilise multi-stage diamond growth to produce a layered structure which in combination with the techniques discussed in this paper will allow for true microdosimetric sized SVs in diamond to be realised.

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5. References
[1] Bauer C et al 1995 Nucl. Instr. Meth. Phys. Res. A 367 207-11
[2] Sato Y et al 2015 Nucl. Instr. Meth. Phys. Res. A 784 147-50
[3] Planskoy B 1980 Phys. Med. Biol. 25 519-32
[4] Yacootts A et al 1990 Phys. Med. Biol. 35 1409-22
[5] Heydarian M et al 1993 Phys. Med. Biol. 38 1035-42
[6] Davis J A et al 2014 IEEE Transactions on Nuclear Science 61 1544-51
[7] Gorjiara T et al 2011 Phys. Med. Biol. 56 4685-99
[8] Rossi H and Zaider M 2012 IEEE Transactions on Nuclear Science 59 3110-6
[9] Davis J A et al 2012 IEEE Transactions on Nuclear Science 59 3110-16
[10] Davis J A et al 2014 IEEE Transactions on Nuclear Science 61 3479-84
[11] Caylar B et al 2013 Appl. Phys. Lett. 103 7-10
[12] Davis J A et al 2017 Appl. Phys. Lett. 110 013503
[13] Marinelli M et al 2001 J. Appl. Phys. 89 1430-5