CHAPTER NINE

GENERAL DISCUSSION AND CONCLUSIONS
9.1 Overview of study objective

Aimed at improving the accuracy of clinical radiation dosimetry using a solid-state detector system, this thesis has explored the radiation detection properties of synthetic diamond crystals of various types and investigated the possibility of developing a multi-purpose synthetic probe for the dosimetry of various beam types under large as well as small-field conditions. The thesis has addressed major challenges usually encountered in the field of diamond radiation detectors in particular and dosimetry in general.

Apart from an improvement in optimising the design and construction of the synthetic diamond probe (section 9.2) for the evaluation of detector performances under clinical conditions, the major findings to be highlighted in this section of the thesis include: (i) the cause of the instability of response of the diamond detectors requiring pre-irradiation; (ii) the influence of various impurity and defect levels on the performances of the diamond sensors; and (iii) the relationship between diamond detector size and field size for measurements under small-field conditions including suitable crystal type and exposure orientation for accurate dose measurements. An overview of the overall dosimetric performance of the presented diamond probe is also discussed and a suggestion for future work is made.

9.2 An optimized synthetic diamond probe for dosimetric applications

Since the purpose of clinical radiation dosimetry is to acquire and evaluate essential beam parameters with a very high degree of accuracy in order to assess the absorbed dose to human tissue, it is relevant to design a radiation detector whose overall performance is nearly independent of various parameters which might influence it. In this thesis, the presented synthetic diamond probe has been optimized for clinical dosimetry i.t.o the following influencing factors: environmental interferences and positional inaccuracies, encapsulation and contact materials.

The diamond probe is unique in its design and construction in that the sensor plate is housed in a holder that is rectangular in shape unlike the usual designs of diamond detectors frequently encountered in literature (Heydarian et al., 1993; van der Merwe et al., 1999; Manfredotti et al., 1998; Marczewska et al., 2007; Assiamah et al., 2007; Lansley et al., 2009; Betzel et al., 2010; Tromson et al., 2010; Schirru et al., 2010; De Angelis et al., 2010) where the diamond plate is fixed in a cylindrical housing. Such a design feature where the probe is aligned in the same orientation as the sensing element eliminates rotational uncertainty in
dosimetry as it ensures ease of positioning the sensor face to the impinging radiation field for accurate dose measurements. Furthermore, the diamond probe is constructed with tissue-equivalent materials with a Perspex body and four carbon-fibre electrodes providing electrical contacts. The probe is additionally provided with features that minimize electromagnetic disturbances such as ambient light which was identified as a factor influencing detector performance (i.e. noisy background and instability of response).

The diamond probe presented here allows crystals to be exposed in different exposure orientations (‘edge-on’ and ‘flat-on’) for impinging radiation without having first to re-orient the diamond sensor within the body of the probe. In addition, the diamond probe is designed to be compatible with commercially available electrometer systems such as PTW Unidos E which is commonly used for dosimetric measurements, unlike other diamond detectors which require specialized electrometer systems such as a Keithley electrometer (Buttar et al., 2000; Ramkumar et al., 2001; Fidanzio et al., 2002; Górka et al., 2008; Tranchant et al., 2008; Betzel et al., 2010; Gervino et al., 2010; Schirru et al., 2010; De Angelis et al., 2010). Also, for free in-air directional dependence measurements, the diamond probe, due to its rectangular shape can be fitted with a circular Perspex build-up cap.

9.3 Rationale for the unstable response of diamond detectors requiring priming
One of the major limitations of the so-called PTW natural diamond detectors was the requirement for daily priming (Bampton, 1976; Vatnisky and Jävinen, 1993; Laub et al., 1999; Björk et al., 2000) and the variation of pre-irradiation dose (between 5 and 15 Gy) between devices due to the presence of uncontrolled amount of impurities and defect levels (Marsolat et al., 2013). This major shortcoming has also been identified in synthetic diamond detectors (Buttar et al., 2000; Fidanzio et al., 2004; Cirrone et., 2006) and it is known that the daily pre-irradiation of diamond detectors is a limiting factor which hinders their suitability as dosimeters, but once primed, the device becomes a suitable detector for dosimetric applications (Whitehead et al., 2001; Guerrero et al., 2004; Bruzzi et al., 2004; Manfredotti, 2005), as the most investigated method to improve diamond detectors is through priming (Manfredotti, 2005).

Furthermore, it has been reported that the priming of some polycrystalline CVD diamond detectors which occurs over several minutes (with higher pre-irradiation doses of up to 30 Gy (Galbiati et al., 2001; Bruzzi et al., 2004; Fidanzio et al., 2004)) compared to natural diamond
detectors is a limitation of CVD diamond detectors over natural diamond and silicon diode detectors (Whitehead et al., 2001). While Buttar et al. (2000) emphasized on the importance to understand and remove the priming effect of CVD diamond detectors in the future, Assiamah (2004) concluded from her study that diamond crystals that require the recommended pre-irradiation step to improve their performances were not suitable to be incorporated into her examined probe for clinical use as attempts to improve the performance of a CVD diamond detector through various methods were unsuccessful. Although Van der Merwe and Keddy (1999) first mentioned that priming was not necessary for their synthetic diamond detector, no investigation as to the reason was undertaken.

Taking note of the role of white light in emptying low level traps in diamond (Burgemeister, 1983) and having observed initially in this thesis that ambient light had a negative effect (i.e. random fluctuations of noisy background) on the performance of the presented probe, the cause of the unstable response of three diamond detectors of various types (HPHT, CVD DG and CVD OG) requiring pre-irradiation was systematically investigated in light and dark conditions by measuring and examining the response of each type of detector as a function of absorbed dose (or progressive X-ray energy). Based on the results of the study, the major cause of the instability of response of the diamond detectors necessitating priming was isolated and attributed to the presence of ambient light which has the effect of emptying various trapping centres present within the diamond sensors.

The percentage difference in response between measurements in light and dark conditions conducted over a period of three consecutive weeks reached values (mean values, averaged over X-ray tube voltage settings between 26 and 30 kVp) of 2.8±1.2, 25.2±6.3 and 63.0±0.3% for the HPHT, CVD DG and CVD OG diamond detectors, respectively. In light conditions alone, the percent change in sensitivity for measurements recorded over two consecutive days were 6.8±1.1, 14.2±1.0 and 14.1±5.7% for the HPHT, DG and OG diamond detectors, respectively. Likewise, for measurements taken only in dark conditions, the percent variation in response between two sets of measurements for the HPHT, DG and OG diamond sensors, respectively were 1.3±0.8, 1.8±0.9 and 2.3±1.6%, which are lower when compared to the percentage values determined for data recorded in light conditions.

The presented results signify that ambient light has a negative influence on the performances of the diamond sensors suggesting therefore that as long as the diamond detectors are
properly shielded from ambient light and their responses stabilised, daily priming is not needed. Furthermore, similar to the finding in literature that some CVD diamond detectors require higher priming doses compared to natural diamond detectors (Whitehead et al., 2001; Galbiati et al., 2001; Bruzzi et al., 2004), the present study demonstrates that the requirement for priming is more significant for the polycrystalline CVD crystals compared to the HPHT diamond as the variation in response under the various measurement conditions is least for the HPHT sample. This could be ascribed to differences in detector quality due to the presence and influence of various defect levels (section 9.4). It is particularly known that polycrystalline CVD films have a high concentration of grain boundaries (Balducci et al., 2005) which accommodate a high level of electron traps (Manfredotti et al., 2006).

9.4  The presence of defects and their influence on the performances of the sensors

Various non-destructive techniques (Raman spectroscopy, ESR, FTIR absorption spectroscopy and TL emission) were utilised in this study to characterise various defect types and/or levels present within each of the diamond samples. A correlation was established between the various defect levels. Firstly, unlike the study presented by Surovtsev et al. (1999) which showed that the Raman-line width increases with increase in [N₃], the results presented in this thesis indicated that there is an inverse correlation between [N₃] and Raman width. Such a finding was related to the observation made by Nam et al. (1991) for HPHT diamonds that crystals synthesized with high levels of N₃ tend to have much lower trap concentrations than those synthesized with lower N₃ levels. While the broadening of the diamond Raman-line width is known to be influenced by crystal defects or non-homogeneous distribution of stress (Pickard et al., 1998; Faggio et al., 1999; Fish et al., 1999), this study established that the broadening in the examined crystals could be mostly linked to defects since the crystals were found to have negligible residual stress values.

Secondly, the concentration of various N-related defects such as the N3 and A/B-centres identified by FTIR absorption spectroscopy decreases with increasing N₃ levels and this was attributed to the sequence of N aggregation in diamond. Thirdly, the TL response values of the crystals (which depict the presence of trapping levels) were found to increase with increasing concentration of defects. The HPHT sample (which should be single crystalline due to its growth method as well as its very narrow Raman width value of 2.27±0.04 cm⁻¹ compared to the broader values (2.34±0.08 cm⁻¹ to 2.60±0.09 cm⁻¹) observed for the CVD crystals) was found to be less defective but with a higher value of N₃ level compared to its
polycrystalline CVD counterparts. This finding could be linked to the combined influences of the non-homogenous structure of polycrystalline materials (Bergonzo et al., 2007), the presence of grain boundaries in polycrystalline CVD diamonds which contain a high concentration of electron traps (Balducci et al., 2005; Manfredotti et al., 2006), the connection between $N_s$ level and trap concentration as observed by Nam et al. (1991), and the sequence of $N$ aggregation in diamond.

The influence of the defect levels on detector performances was subsequently investigated within an optimized probe. Initially, mammography X-rays were employed to investigate the cause of the unstable response of various diamond detectors requiring the often cited pre-irradiation step (chapter 5). Mammography X-rays were chosen to serve as a possible tool to probe the presence, distribution and effects of trapping levels of varying depths with changes in X-ray energy. The overall better performance (i.t.o stability of response or priming requirement) of the HPHT diamond sensor compared to the CVD sensors is attributed to the presence of fewer defects or shallow traps within the HPHT sample as evidenced by the results of the characterization techniques.

Subsequent to the initial study, the influence of the defect levels on the dose rate linearity index $\Delta$ was systematically examined (chapters 6 and 7) due to its influential role in dosimetry. The $\Delta$ values of the crystals (computed on exposing the crystals to high-energy electron and photon beams) were also evaluated for their dependence on bias voltage and beam energy. Compared to a number of studies which concluded that $\Delta$ is independent of bias voltage (Planskoy, 1980), beam energy and type (Planskoy, 1980; Laub et al., 1999; Fidanzio et al., 2004; Cirrone et al., 2006; Björk et al., 2000), this thesis established that such a conclusion can not be generalised. The $\Delta$ values of the diamond sensors were found to vary with these parameters, and a change of $\Delta$ with defect levels was observed, indicating that differences in crystal quality due to the presence of various defects could alter the value of $\Delta$.

The study demonstrated that the $\Delta$ values of crystals with high defect levels differed strongly with bias voltage and electron energy compared to photon energy, and it was established that the change in $\Delta$ with bias voltage would be within 2% when the TL response value is $\leq$2.2 arbitrary units. Further investigation suggested that the combined effect of the N3 and C-H defect centres could have a dominant influence on the overall dependence of $\Delta$ on bias voltage and beam energy. The percent difference of $\Delta$ between two beam energies of the
same type (electrons or photons) was found to increase with increasing concentration of the N3 and C-H centres, and it was observed that within a given range for the N3 defect levels the deviation of Δ between two energies of each beam type would be within 2%. A correlation between TL response and each of these two defect levels was established. The stronger variation of Δ with electron energy compared to photon energy was attributed to the greater influence of surface defects such as the C-H defect centres.

Unlike the study presented by Marczewska et al. (2007) which highlighted that (evaluated under 60Co radiation) varied with nitrogen concentration though it was not obvious in their study which type of nitrogen impurity was causing the variation, this thesis has isolated pertinent defect types which could influence the change of Δ. As a correlation between TL response and each of the N3 and C-H defect levels was observed, this study demonstrates that TL emission could be a valuable parameter in probing the performances of diamond crystals. Mavunda et al. (2008) also concluded in their study that TL may be a tool to consider for the selection of CVD diamond crystals for general alpha spectroscopy.

9.5 Suitable crystal type, size and orientation for measurements under small fields

With rapid advancements in technology, the application of high-energy teletherapy procedures in the future could be shifting towards the implementation of small-field techniques. Advanced techniques such as IMRT utilize small radiation fields (Zhu, 2010) so as to spare normal healthy tissues while lethal radiation doses can be delivered to tumour volumes (Barnett et al., 2005; Das, 2009; Marsolat et al., 2013). Consequently, dosimetry in the future may rely on measurements in small fields.

For measurements under small-field conditions, three vital parameters are required of radiation dosimeters: high spatial resolution; high radiation sensitivity; and tissue-equivalence of the detector material and its housing. The requirement for high spatial resolution, which is dependent on detector size or sensing volume, is necessary in order to reduce the so-called volume effect of detectors which become significant in small fields due to steep dose gradients. Tissue-equivalence of the detector is required so as to reduce perturbation effects and local changes in charged particle equilibrium (Somigliana et al., 1999; Westermark et al., 2000; Haryanto et al., 2002; Laub and Wong, 2003; Scott et al., 2012), and this requirement is dependent on the atomic composition of the detector material and its encapsulation.
High sensitivity of the detector to radiation is essential to keep statistical noise and measurement time within an acceptable level (Westermark et al., 2000). The necessity for high radiation sensitivity also implies that a detector with small physical size can be utilised to meet the requirement of high spatial resolution. As the requirement for high radiation sensitivity is dependent on the SNR (Manfredotti et al., 1998), it is essential to select a detector material with a very low background since leakage currents and their fluctuations are an undesirable noise source in radiation measurements (Kania, 1993) and a limiting factor for charge collection efficiency (Fidanzio et al., 2002; Fidanzio et al., 2004). The better the SNR, the faster the measurements can be conducted, and this property is suitable for measurements of relative beam data such as beam profiles, PDD curves and TMRs.

Given the requirement of a higher accuracy for small-field dosimetry and based on the results of electrical and various dosimetric characterizations presented in this thesis, the HPHT diamond sensor seems to be a suitable detector material for measurements under small-field conditions when compared to its polycrystalline CVD counterparts. Firstly, the HPHT diamond was found to have the lowest and most stable leakage current. Secondly, the results of the systematic investigation into the cause of the instability of response of the three types of the diamond detectors (examined in this thesis) requiring priming established that the HPHT diamond had an overall better performance. Thirdly, a systematic study of the dose rate dependence of the various diamond detectors demonstrated that only the values of the HPHT diamond did not vary with beam energy. Fourthly, characterizations of the diamond detectors under small-photon-field conditions illustrated that the HPHT sensors due to their low defect levels performed better than the CVD crystals.

It was established from measurements under small-field conditions that the dose difference between the OFs measured with the various detectors and a small-field detector (Diode E) would be within 3% when the detector size is not greater than 3/4 of the field size. Furthermore, the presentation indicated that the ‘edge-on’ exposure orientation of a diamond detector due to its greater attenuation depth and reduced surface area appears to be a suitable geometry for OF measurements especially for very small fields (< 1x1 cm²). For example, a maximum dose deviation of 1.9% was observed between the OFs measured with Diode E and a small-sized HPHT diamond sensor of 9 mm³ sensing volume in the ‘edge-on’ orientation compared to a 4.6% deviation in the ‘flat-on’ geometry down to a 0.4x0.4 cm² field.
9.6 Overall dosimetric performance of the prototype synthetic diamond probe

There have been significant improvements in the field of medical dosimetry with synthetic diamond detectors. Though often met with various challenges, most reports in literature have characterized the dosimetric performance of a synthetic diamond detector to either low-energy X-rays (Assiamah et al., 2007), orthovoltage therapy X-rays (Buttar et al., 2000; Ramkumar et al., 2001), high-energy electron beams (Van der Merwe and Keddy, 1999), high-energy photon beams (Fidanzio et al., 2002; Marczewska et al., 2007; Górka et al., 2008; Tranchant et al., 2008; Betzel et al., 2010; Gervino et al., 2010; Schirru et al., 2010; De Angelis et al., 2010; Ciacaglioni et al., 2012; Marsolat et al., 2013) or both high-energy photons and electrons usually using one energy of each beam type (Fidanzio et al., 2004; Bruzzi et al., 2000) unlike the present study which has evaluated detector performances within an optimized probe on exposure to low-energy X-rays, high-energy electron and photon beams.

Various parameters were used to characterize detector performance. These include but not limited to: relative beam data such as electron PDD, OFs for both electron and photon beams and $^{60}$Co TMRs; linearity measurements; directional and dose rate dependence measurements, radiation sensitivity per unit detector sensing volume and SNR. The ‘edge-on’ orientation of the detecting element (i.e., diamond sensor) of the probe, due to its greater attenuation depth and small surface area was found suitable for low-energy X-ray radiation dose measurements while the ‘flat-on’ geometry was established to be more appropriate for electron PDD profiling due to its large and flat surface area. Because of the high penetration power of high-energy photons, both exposure orientations were determined to be suitable for dose measurements. However, for the measurements of output factors in very small field sizes (below 1x1 cm$^2$), the ‘edge-on’ orientation was found to be more appropriate. Thus the prototype synthetic probe presented in this thesis is a multi-purpose radiation detector as it could be used for the dosimetry of various beam types.

A systematic study of the influence of Δ in relative dosimetry indicated that with a suitable diamond sensor, relative distributions measured with the diamond probe would compare favourably with those obtained with reference dosimeters in the order of 1–2% with or without dose rate dependence corrections. Such a performance was attributed to the negligible energy dependence of the diamond probe and its minimal and stable background signal. It was further demonstrated that even if a crystal shows significant dose rate
dependence, appropriate corrections could, if needed, be applied to its response. Additionally, it was established that at a given depth of measurement a single value could be used for various beam energies within a given clinical range if the variation of with beam energy is within 2%. The justification of this level of accuracy when the dose rate is varied by a factor of 10 is provided in Appendix B.

Given the importance of the angular response of a detector in an electron field where the distribution in energy and direction of electrons changes quickly with depth of penetration, it is relevant to use detectors which are independent of energy and beam direction (Brahme and Svensson, 1976; Heydarin et al., 1993; Björk et al., 2000). Employing two diamond sensors (a HPHT sample and a CVD DG diamond), the angular dependence of the synthetic diamond probe was evaluated between 0° and 105° on exposure to electron beams using an applicator defined field size of 10x10 cm². Between 0° and 60°, the response remained flat within 2%, whereas a stronger dependence of about 3.7% was observed between 60° and 105° for both detectors. These results compare favourably with those reported by Ciancaglioni et al. (2012) for a synthetic single crystal diamond detector where a maximum variation in angular response of 3.5% was established in a 3x3 cm² field size in order to reduce possible spurious noise that could arise from cable irradiation.

It is known that the directional response of solid state detectors is due to changing interface effects (Rikner, 1985; Heydarian et al., 1993) where a varying degree of secondary electron spectrum equilibrium is established within the sensing volume of a detector (ICRU, 1972). In this study, the 2% variation in angular response observed between 0° and 60° was related to the interface phenomenon, whereas the stronger dependence of about 3.7% established beyond 60° was ascribed to a geometry effect. This suggests that the mechanical design of a radiation detector could influence its angular response. Further investigation then indicated that the performance of the diamond probe could be improved by incorporating a jig for the response to be independent of incident beam direction.

As the linear response of a diamond detector with absorbed dose and dose rate are primary requirements in radiotherapy (Marczewska et al., 2007), an evaluation of the performance of the diamond probe illustrated that its response was linear with absorbed dose and time on exposure to electron beams and to ⁶⁰Co radiation, respectively. On exposure to
mammography X-rays, the response of the diamond probe was linear with tube loading and tube voltage which are two parameters that determine patient dose in mammography.

Due to the importance of high SNR and radiation sensitivity for measurements under small-field conditions, values of about $5.5 \times 10^3$ and 197.3 nC Gy$^{-1}$ mm$^{-3}$ for the SNR and sensitivity, respectively were established for the probe, under 6 MV photon irradiation using a HPHT diamond (HPHT2) as sensor in a 0.4x0.4 cm$^2$ field. In comparison, a sensitivity value of 136.1 nC Gy$^{-1}$ mm$^{-3}$ was obtained with a small-field diode detector in the same radiation field. Results for such field sizes have not been reported in literature for other diamond detectors. With the same HPHT diamond probe in a 10x10 cm$^2$ 6 MV photon radiation field, a SNR of $1.8 \times 10^4$ was obtained compared to the values of 10 and $2.6 \times 10^3$ reported for polycrystalline (Fidanzio et al., 2004) and single crystal CVD diamond detectors (Tromson et al., 2010) respectively, under the same conditions. Also, a sensitivity value of 652.2 nC Gy$^{-1}$ mm$^{-3}$ was established for the diamond probe in the 10x10 cm$^2$ radiation field compared to a value of 0.72 nC Gy$^{-1}$ ($\approx 189.5$ nC Gy$^{-1}$ mm$^{-3}$) determined for a synthetic single crystal diamond detector in a Schottky diode configuration (Ciancaglioni et al., 2012). Furthermore, sensitivity values of 20–135 nC Gy$^{-1}$ mm$^{-3}$ have been reported for other synthetic and PTW diamond detectors in a 10x10 cm$^2$ field using a 6 MV photon beam (Fidanzio et al., 2002; Fidanzio et al., 2004).

On exposure to an electron beam using an applicator defined field of size 10x10 cm$^2$, a sensitivity value of 547.52 nC Gy$^{-1}$ mm$^{-3}$ was also established for the probe using a HPHT diamond crystal (HPHT1) compared to the value of 91 nC Gy$^{-1}$ mm$^{-3}$ reported for a synthetic diamond detector (Bruzzi et al., 2000).

9.7 Conclusions

The major findings established in this thesis through systematic investigations include:

(i) Differences in crystal quality due to the presence and influence of various defects cause Fowler’s dose rate linearity index, $\Delta$, to differ strongly with bias voltage, beam energy and type. Apart from TL which has been identified as a suitable tool to probe the performances of the examined crystals, a number of defects such as N$_a$ impurities, N3 and C-H centres were isolated as pertinent defect types that could strongly influence the dosimetric performances (i.t.o) of diamond sensors suggesting that diamond crystals could be selected or perhaps
tailor-made with various impurity levels which when used as radiation sensors for dosimetric applications display optimum performance;

(ii) The primary cause of the instability response of the examined diamond sensors requiring the often proposed pre-irradiation step prior to each measurement was isolated and attributed to the presence and influence of ambient light. Such a finding implies that once an appropriate diamond crystal has been selected and coupled to the probe presented in this thesis with the diamond probe properly shielded from ambient light and its response stabilised, daily priming is not necessary.

(iii) Based on experimental evidence for measurements under small-photon-field conditions, it can be formulated as a rule of thumb that if diamond detector size is \( \leq 3/4 \) of field size, an accuracy level within 3% could be achieved for relative dose measurements with the ‘edge-on’ orientation being a suitable detector geometry in very small field sizes (below 1x1 cm\(^2\)). It was also demonstrated that with a selected diamond sensor, the dosimetric performance of the diamond probe on exposure to various teletherapy beams would compare favourably with that of a reference dosimeter in the order of 1–2% with or without dose rate dependence corrections. In addition, a higher figure of 197.3 nC Gy\(^{-1}\) mm\(^{-3}\) detector sensitivity per unit sensing volume was established compared to a value of 136.1 nC Gy\(^{-1}\) mm\(^{-3}\) obtained with a small-field diode detector;

(iv) Overall, a HPHT diamond sensor, due to its low defect levels was found to perform better (i.t.o low and stable background signal, measurements under small-field conditions, stability of response (or priming requirement), and the independence of its \( \Delta \) values on bias voltage, beam energy and type) compared to the CVD crystals. One CVD diamond crystal (DGB1) was also found to be more suitable as a sensing element compared to others.

The results of various dosimetric characterizations do indicate that once a crystal is chosen and coupled to the probe, then the near-tissue equivalent diamond probe could be utilised for clinical applications in large as well as small radiation fields. Its potential use for clinical dosimetry was demonstrated by its linear response characteristics with absorbed dose, dose rate and exposure parameters to various beam types, negligible energy dependence and a small variation in angular response, high radiation sensitivity and SNR, minimal and stable dark current, and the added advantage not to require daily pre-irradiation if properly shielded.
from electromagnetic interferences. Furthermore, relative dose distributions measured with the diamond probe were found to be in close agreement with energy-corrected data obtained with reference ion chambers.

9.8 Recommendation for future work

Due to the correlation observed between the \( \Delta \) values of the diamond sensors and the various defect types and levels identified in this thesis, a study aimed at an understanding of the role of the defects on the sensitivity values of the diamond crystals is suggested as future work.