**Diamond x-ray dosimeters**

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## A. Introduction

The interest in the use of diamond as radiation dosimeter stems principally from its tissue equivalence. The atomic number of diamond (Z=6) matches that of the biological tissue being exposed to high energy photon radiation (the effective number of human tissue is 7.42 for muscle and 5.92 for fat), therefore the energy deposited by any beam of radiation for unit of mass will be virtually the same for tissue and for diamond. This fact makes diamond detectors more suitable than any other solid state dosimeter, e.g. the commonly used silicon diodes, for photon energies below 200 keV where the absorbed energy depends at least from the fourth power of the atomic number.

Other attractive properties of diamond as dosimeter are that it is chemically stable, non toxic, mechanically robust and relatively insensitive to radiation damage. Diamond detectors can be fabricated with small volumes (of the order of 1 $mm^3$ ) and then can be considered for in vivo dose measurements. As a consequence, diamond is particularly suitable for accurate dose measurements in small radiation fields such as in stereotactic radiosurgical beams where ion chambers are inappropriate because of their poor spatial resolution and their sensitivity to lateral electronic disequilibrium [1]

Furthermore, diamond is an extremely versatile material and its dosimetric properties can be exploiting as a thermoluminescence (TL) or thermoconductive (TSC) dosimeter, as a solid state ionisation chamber (IC) and as a radiophotoluminescence detector [2][3].

So far, commercial diamond detectors are fabricated by natural gems. The main drawbacks of these dosimeters are the high cost and long waiting time [4], due to the severe selection of gems with suitably quality, and the poor reproducibility overall in the case of TL dosimeters. However, the recent availability of synthetic samples, grown under controlled conditions both by the High Pressure-High Temperature (HPHT) and Chemical Vapour Deposition (CVD) techniques make in perspective possible a widespread application of diamond in medical radiation dosimetry.

## B. As an ionisation chamber

Almost all the works published up to now describe the dosimetric properties of pure IIa natural diamond solid state ionisation chambers [1][3][4][5][6][7][8][9][10][11][12][13][14][15]. Kozlov et al. [5] has summarised the severe criteria to select gems suitable as radiation detectors: gems should have high resistivity ($\geq 10^{12}$ $\Omega \cdot$cm) and low impurity content (nitrogen should be less than $10^{19}$

atoms/cm$^3$). The latter requirement is particularly important to guarantee a recombination rate almost independent from the electron/hole generation, i.e. an almost linear increase of photocurrent with dose rate and an almost complete charge collection. Furthermore, charge carriers in traps form a space charge which gives rise to a counter electric field which opposes to the applied bias voltage (polarisation effect). Kozlov et al. [5] first demonstrated that the use of special injecting contacts (e.g. boron implanted contacts), greatly contribute to neutralise such space charge effects by a significant increase of dark current. Using contacts with different injecting properties, the linearity of the photoresponse can be extended down to few mR·s$^{-1}$ (about $10^{-5}$ Gy·s$^{-1}$), even if a significant increase of dose rate threshold due to the increase of dark current has to be taken into account. Another commonly used method to maximise the detector response and to stabilise the current consists in a strong pre-irradiation (or priming) of the diamond detector with doses ranging from 1 to 10 Gy. This strongly recommended pre-.treatment is essential to fill charge traps and to reach an equilibrium trap population which allows a stable response of the detector to be reached and settled for long times [10].

Diamond detectors show a response that is in general slightly decreasing with dose rate. The empirical expression of the dependence of the photocurrent $I_{ph}$ with dose rate $\dot{D}$ is

(1) $I_{ph} = I_0 + S \cdot (\dot{D})^{\Delta}$

where $I_0$ is the dark current; $\Delta$ and S are the phenomenological parameters indicating the slight sublinearity of the response and the sensitivity, respectively. Some values of $\Delta$ obtained by various authors are listed in table I. $\Delta$ ranges between 0.9 and 1, is to be considered almost constant, is very slightly dependent on photon beam energy [4], and has to be considered as peculiar of the material.

The sensitivity S is calculated by the photocurrent/(dose rate) ratio where the denominator is evaluated by means of the exposure rate $\dot{X}$ measured by an ionisation chamber (IC) taking into account the mass energy absorption coefficient of carbon and air shown in fig. 1.

(2) $\dot{D} = \frac{W}{e} \cdot \frac{(\mu_{en}/\rho)_C}{(\mu_{en}/\rho)_{air}} \cdot \dot{X}$

where $\dot{D}$ and $\dot{X}$ are expressed in terms of SI units (Gy/s and C/(kg·s), respectively) and (W/e) is the mean energy expended in air per ion pair formed and per electron charge (W/e=(33.97±0.06)J/C [16])

Carbon is nearly water equivalent and, as a consequence, the diamond response is nearly directly proportional to the absorbed dose rate of water. The mass energy absorption coefficients ($\mu_{en}/\rho$) of carbon, air, silicon and LiF relative to water as a function of photon energy are shown in fig. 2. The assumption of energy independent correction factors to convert diamond response to dose, involves

a systematic error of less than 7% to be compared with the changes of more than 40% relevant to silicon to water ratio [15].

The mean carrier lifetime $\tau$ can then be evaluated [7] by assuming a simple photoconductive mechanism, fully depleted detectors, ohmic contacts and 13 eV as the mean energy to generate electron/hole pairs [6]. Apart the result obtained by Burgemeister [7] , all the values of $\tau$ relevant to natural diamond lie between 1-10 ns as stated by Kozlov et al. [5]. Burgemeister [7] obtained exceptionally high results for one small, very accurately selected IIa diamond sample which showed a photoconductive gain larger than 100.

Very little information is currently available synthetic diamonds used as solid state ionisation chambers. Keddy and Nam [11] showed that diamond samples with nitrogen content around 100 ppm synthesised by the HPHT method have responses both to alpha particle and to gamma ray radiation comparable with natural gems.

High quality CVD diamond detectors have been recently fabricated[17][18]. These preliminary results demonstrate that CVD diamond detectors show dosimetric properties close to those measured in natural diamond but with a much higher reproducibility and lower costs. Moreover, the fabrication of microdosimeters realised by the deposition of thin diamond films on metallic tips makes realistic the possibility of making "pin-point" detectors [3] with sensitivity/mass ratio close to ionising chambers, for special treatment, as in brachytheray [13]and stereotactic radiosurgery [14], where high spatial resolution is required.

**B. As a thermoluminescence or thermoconductive detector**

Thermoluminescence is the thermally stimulated emission of light from an insulator or a semiconductor following the previous absorption of energy from radiation [19].

Some of the charge carriers generated by absorbed ionising radiation remain trapped in discrete energy levels within the forbidden energy gap due to impurities or lattice imperfections. Subsequent heating of the material can release the trapped electrons/holes, which can recombine radiatively. The energy of the emitted photons is then peculiar of the material, or, better, of the trap level distribution, whereas the TL intensity is function of the trapping cross section, temperature, trap density and absorbed energy of radiation. This latter feature, as well as the simple and relatively inexpensive experimental set-up, makes TL a widely used technique for detecting ultraviolet and ionising radiation in the area of health physics, radiation protection, space sciences and biomedical sciences.

Although diamond was probably the first solid in which TL was observed (Robert Boyle 1663), very few paper have been published on natural diamond TL properties because of its poor reproducibility and low signal [20][21].

Nam and Keddy [22], first studied the performances of HPHT diamonds with controlled amount of impurities. They showed that the TL sensitivity of diamonds with low nitrogen content (less than 10 ppm) is over three order of magnitudes higher than in gems with 100 ppm of nitrogen. Boron content of the order of 1 ppm increases the linearity from less than two decades to over three decades, whereas higher concentrations introduce supralinearity into the TL response. They showed that a controlled introduction of boron which partially compensates the nitrogen content allows diamond thermoluminescent dosimeters to be fabricated with a linearity of response extending from 0.01 to 10 Gy, sensitivities equivalent to LiF and a high temperature component with an half life of approximately 200 days [3].

Several papers have been published in the last five years on the use of diamond TL dosimeters synthesised by the CVD technique under controlled conditions.

In table II some features of CVD diamond dosimeters are summarised. All the measurements were carried out using beta particles from a $^{90}Sr$ excitation source. A pre-heating (PH) process was also used to suppress the unstable low temperature TL peaks and to isolate the high temperature structures (PT).

The kinetics of the TL process was studied both by deconvolution of glow curves [20][21][23][24], isothermal decay [21][24] or initial rise method [20]. It is worth noticing that almost all the authors obtained the best fit of the glow curves by using first order kinetics equations, i.e. assuming a model in which one trap-one recombination centres are present without retrapping. The main temperature TL peak can be reasonably ascribed to the presence of a level at around 1 eV. Boron doped CVD diamonds exhibits glow curves that have been interpreted by assuming a continuous trap level distribution and more complex kinetic process [23].

Furetta et al. [25] observed no fading for a period of one month for one sample stored in a Pb box at a constant (21°C) temperature. Optical bleaching has been reported by Vittone et al. [24] if the sample is illuminated with light in the spectral range 450-550 nm. A linearity range extending up to 20 Gy and a flat energy response for x-ray energies between 0.1 and 1.25 MeV is reported by Furetta et al. [25]. Comparative studies of the TL response to beta irradiation of diamond and LiF dosimeters have demonstrate comparable sensitivities of the two materials [20][26]. The spectrum of light emission shows a broad band centred at 498 nm [20]. If such a spectral feature is peculiar of the TL mechanism and valid for all the samples, a large margin of improvement in TL sensitivity can be foreseen by using photocathodes with more suitable quantum efficiency spectral response.

Other investigations have been recently carried out to gain better understanding of band gap states. In particular, thermally stimulated current technique was recently used for trap level spectroscopy and for UV dosimetry[27][28] .

### C. As a radiophotoluminescence detector

Araikum et al. [29] studied the properties of diamond as a radiophotoluminescence dosimeter. The sample was first exposed to ionising radiation and then subjected to de-excitation illumination (usually UV). The intensity of the photoluminescence peak (526 nm) was directly proportional to the absorbed dose in the range from 0.1 to 10 kGy. Possible applications of such dosimeters can be found in sterilisation by radiation fields [3].

### D. Conclusions

Diamond is an ideal candidate as a radiation detector since it is chemically stable, inert and essentially soft tissue equivalent. Moreover, diamond is an extremely versatile material and its application as a detector for ionising radiation can be considered in various modalities. Some key parameters of the performances of diamond dosimeters as measured and reported by different authors are summarised in this datareview.

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[32] SCANDITRONICS Therados silicon semiconductor GR-p detector

[33] Wellhofer Dosimetrie IC10 Ionisation Chamber..

[34]

| Detector | T (mm) | V ($mm^3$) | S ($C \cdot Gy^{-1}$) | S/M ($C \cdot J^{-1}$) | τ (ns) | $I_0$ (pA) | □ |
|---|---|---|---|---|---|---|---|
| Natural diamond [5] D-10 | 0.4 | 7.6 | $2.6 \cdot 10^{-6}$ [1250 keV] | 0.01 | 3.5 | 0.04 [300] | |
| Natural diamond [6] C1 | 1 | 3 | $0.06 \cdot 10^{-6}$ [661 keV] | 0.006 | 2.6 | 1 [150] | 0.91 |
| Natural diamond [7] | 1 | 0.8 | $31 \cdot 10^{-6}$ [≈1000 keV] | 11 | 14700 | ≤0.6 [50] | |
| Natural diamond [8] | 0.33 | 1.4 | $0.175 \cdot 10^{-6}$ [1250 keV] | 0.037 | 2.6 | ≤1 [100] | 0.98 |
| Natural diamond [9] | (0.2-0.3) | ≈3 | $\approx 0.5 \cdot 10^{-6}$ [1250 keV] | ≈0.05 | 1.9 | ≤1 [100] | 1 |
| Natural diamond [5] [4] | 0.32 | 3 | $0.41 \cdot 10^{-6}$ [4-20 MV photon beam] | ≈0.04 | 2.6 | ≤3 [100] | 0.963 |
| Natural diamond [15] | 0.26 | 1.9 | $0.22 \cdot 10^{-6}$ | 0.03 | 1.4 | | |
| HPHT diamond [11] | | | $0.01 \cdot 10^{-6}$ [1250 keV] | | | ≤2.5 [60] | |
| CVD diamond [17] | 0.15 | 210 | $8.1 \cdot 10^{-6}$ [1250 keV] | 0.01 | 0.1 | 10 [150] | |
| CVD diamond [18] | 0.4 | 24.8 | $2.7 \cdot 10^{-6}$ [6 MV photon beam] | ≈0.03 | 3.2 | 50 [100] | |
| CVD diamond tips [18] | ≈0.02 | $<3 \cdot 10^{-2}$ | $0.007 \cdot 10^{-6}$ [6 MV photon beam] | 0.07 | | 1 [100] | |
| Silicon diode [32] | 0.06 | 0.4 | $0.21 \cdot 10^{-6}$ [20 MeV electron beam] | ≈0.15 | | | |
| IC [33] | | 144 | 0.005* [1250 keV] | | | | |

Table I: dosimetric properties of diamond detectors working as ionisation chambers. T=thickness, V=active volume, S=sensitivity [radiation energy], S/M=Sensitivity to mass ratio, $I_0$=dark current [at operating bias voltage]. Lifetime τ is evaluated considering a mean carrier mobility of 2000 $V \cdot s^{-1} \cdot cm^{-2}$ and a mean energy to create an electron/hole pair of 13 eV [31], Δ is the sublinear exponent. *Absorbed dose to water

| Detector | Excitation source | PH (K) | □ (K·s$^{-}$ | PT (K) | E (eV) | s (Hz) | b |
|---|---|---|---|---|---|---|---|
| CVD diamond [20]-TL | $^{90}$Sr | 500 | 0.5 | 540 | 0.86±0.07 | $4.2\cdot10^{6}$ | 1 |
| CVD diamond [21]-TL | $^{90}$Sr | 573 (1 h) | 2 | 550-560 | 1.275<br>0.646 | $4.79\cdot10^{9}$<br>$2.85\cdot10^{4}$ | 1 |
| CVD diamond [24]-TL | $^{90}$Sr | 473 (30 s) | 8 | 513 | 1.1 | $4.6\cdot10^{10}$ | 1 |
| CVD diamond [26]-TL | $^{90}$Sr | 493 (3 min) | 2 | 571 | 0.92 | $2.92\cdot10^{7}$ | 1 |
| Boron doped CVD diamond [23]-TL | $^{90}$Sr | | 2 | 377<br>442<br>492<br>525 | 0.62<br>0.75<br>1.13<br>1.64 | $7.1\cdot10^{7}$<br><br><br>$3.3\cdot10^{12}$ | 1.6<br>1.4<br>1.1<br>1.2 |
| CVD diamond [27]-TSC | Deuterium lamp | 543 (10 min) | ≈0.2 | 555 | 1.86±0.05 | $3\cdot10^{14}$ | 1 |
| CVD diamond [28]-TSC | Deuterium lamp | | ≈0.2 | 520 | 1.42<br>0.87 | | 1 |
| IIa diamond [28]-TSC | Deuterium lamp | | ≈0.2 | 330 | 0.68<br>0.78 | | 1 |

Table II: dosimetric properties of diamond samples working as thermoluminescence or thermoconductive dosimeters. PH=Pre-heating temperature, β =heating rate, PT= peak temperature, E=activation energy, s=frequency factor,, b=kinetic order.

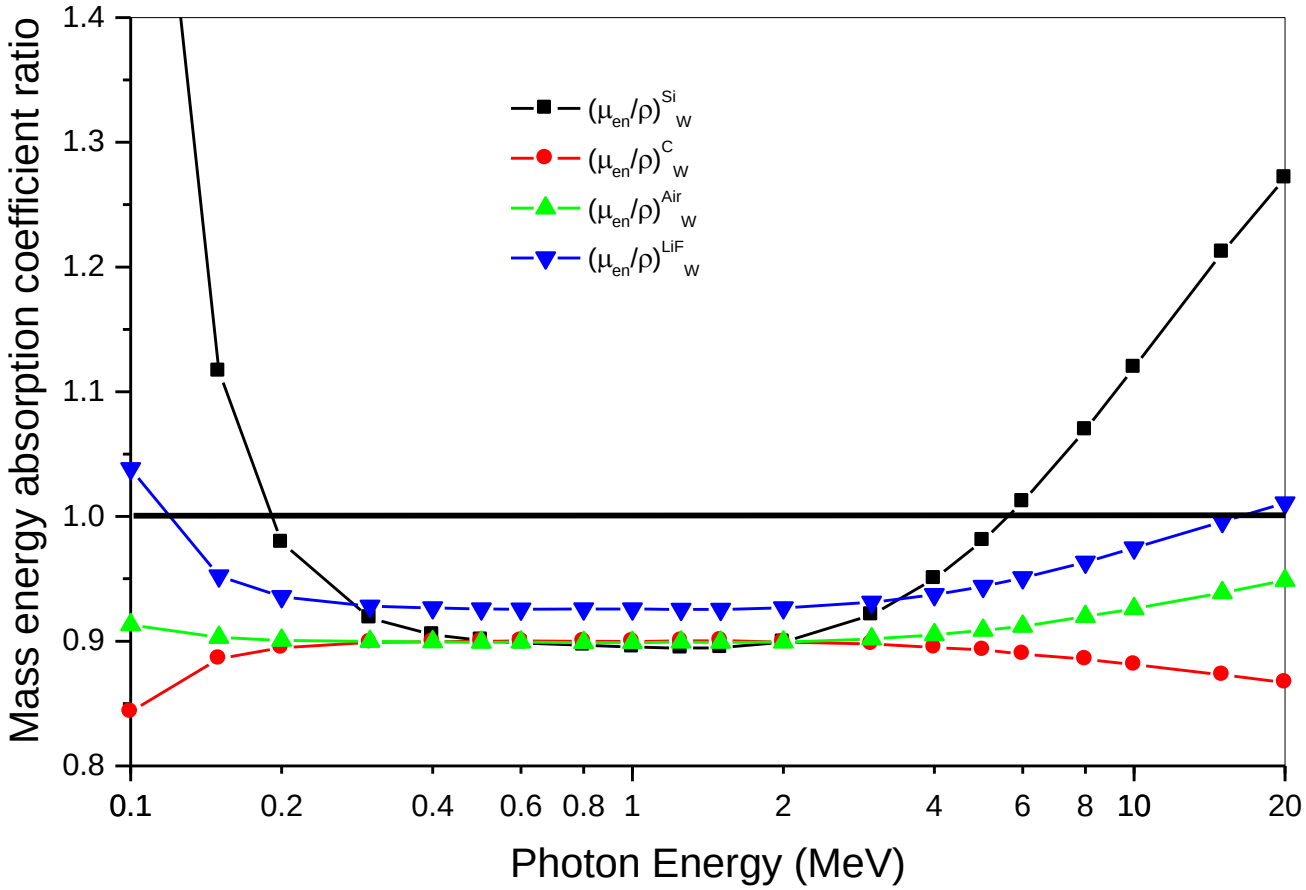


Fig. 2: Mass energy attenuation coefficients $\left(\mu_{en}/\rho\right)_{w}^{xxxx} = \frac{\left(\mu_{en}/\rho\right)_{xxxx}}{\left(\mu_{en}/\rho\right)_{water}}$ (xxxx=C,air,Si,LiF ) relative to water as a function of photon energy [30]

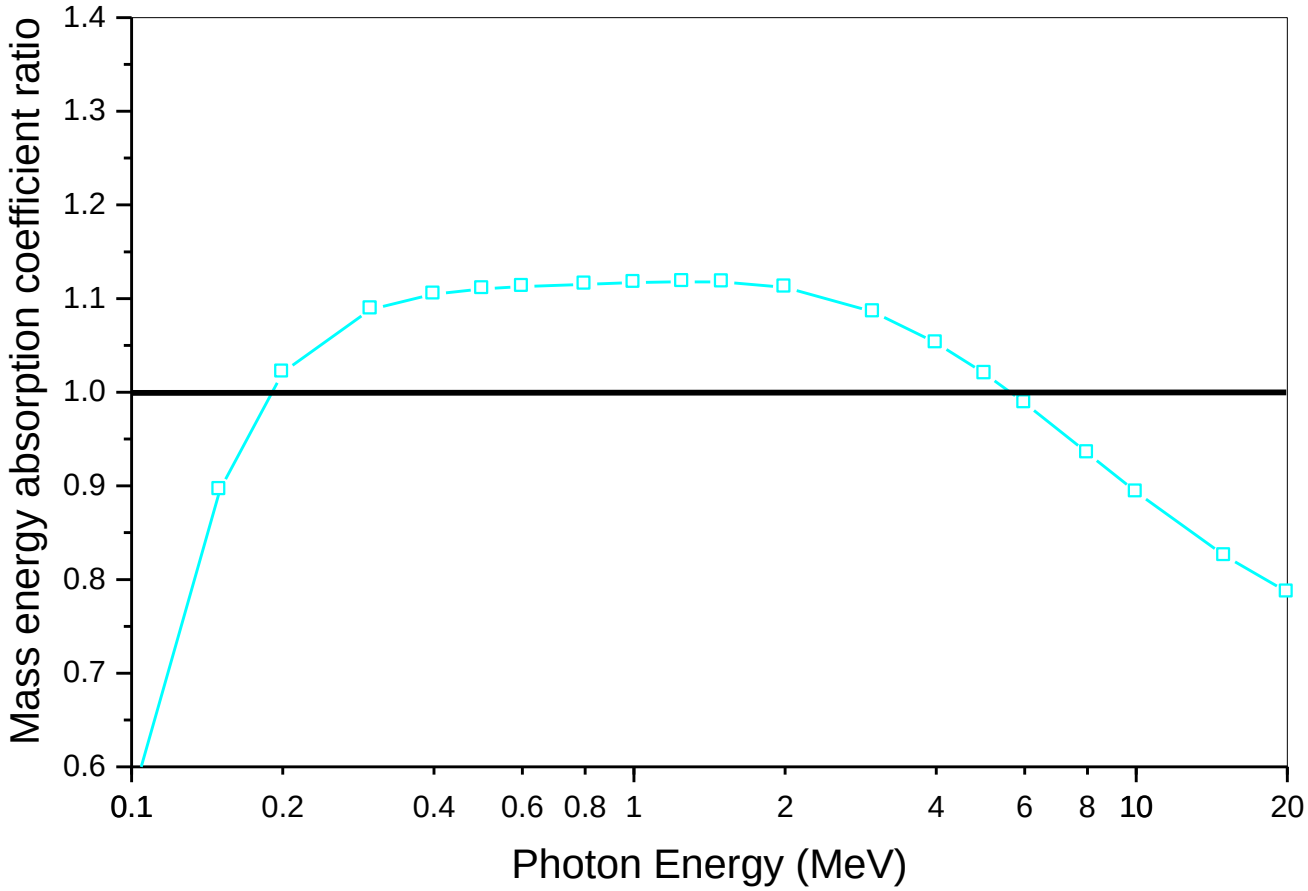


Fig. 1: Mass energy attenuation coefficients $\frac{\left(\mu_{en}/\rho\right)_C}{\left(\mu_{en}/\rho\right)_{airr}}$ of carbon relative to air as a function of photon energy [30].