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Toward the use of single crystal diamond based detector for ion-beam therapy microdosimetry

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A B S T R A C T

In this work, the fabrication and characterization of a microdosimeter based on a synthetic single crystal diamond is reported. The microdosimeter is realized by means of both standard photolithography and selective chemical vapor deposition techniques to accurately define its micrometric sensitive volume.

Experimental measurements were carried out at the Ruđer Bošković Institute microbeam facility using different particles such as proton, helium, lithium, carbon and oxygen. Ion beam induced charge (IBIC) technique was performed to characterize the microdosimeter response in terms of its charge collection properties. The experimental data were also analyzed by means of Geant4 Monte Carlo simulations.

Diamond based microdosimeter shows a well-defined active volume. Homogeneity of the response was estimated at about 7% and linked to structural defects of the diamond surface as deduced by AFM inspection. The detector response shows a good linear behavior for ions of different kind and energy, indicating that the detector is suitable for measuring a wide range of particles and LET i.e. 100 ÷ 3000 keV/μm. Finally, microdosimetric capabilities of the diamond based microdosimeter were preliminarily tested in low LET radiation fields (i.e. protons beam).

1. Introduction

Diamond has been identified as a desirable material for use in microdosimetry due to its near tissue equivalence for dosimetry of photon fields (its atomic number, Z = 6, is very close to Z_{eff} = 7.5 for soft tissue) and radiation hardness (Nemanich et al., 2014; Sussmann, 2009; Pillon et al., 2008). Furthermore, Monte Carlo simulations indicate that microdosimetric energy deposition spectra in diamond can be converted to equivalent energy deposition spectra in water by applying a simple scaling factor, for protons of energy ranging between a few MeV and 250 MeV (Davis et al., 2014a).

A microdosimeter should be able to measure the distribution of imparted energies in targets of micrometric sizes exposed to complex radiation fields, providing the information that, together with absorbed dose, relates to radiobiological effects (RBE) of ionizing radiation (Wilson, 1946; International Commission on Radiation Units and Measurements, 1983). Microdosimeters based on chemical vapor deposition (CVD) single-crystal diamond have been recently proposed in the literature (Davis et al., 2014b, 2017; Rollet et al., 2012; Verona et al., 2015) for ion-beam therapy applications. In particular, diamond based Schottky diodes have been fabricated and tested in the framework of a collaboration between "Tor Vergata" University and EBG MedAustron (Verona et al., 2015; Magrin et al., 2015) to assess their possible use during the phases of commissioning of the ion beams and the clinical periodic routines. These detectors exhibit several advantages such as zero bias voltage operation, small dimension, good spectroscopic properties (i.e. charge collection efficiency close to 100% and good energy resolution) and high radiation tolerance (Pillon et al., 2008; Verona et al., 2015).

Such devices could be useful for other radiation therapy applications too. Targeted alpha-radionuclide therapy (TAT) employs radiopharmaceuticals labelled with alpha-emitters to deliver high dose to tumor cells with high specificity (Sgouros et al, 2010). Many radiopharmaceuticals for TAT are currently undergoing different phase trials, however their clinical implementation would definitely benefit from a better understanding of the RBE for different cell lines and endpoints. The micro-metric thickness of the sensitive region of diamond detector compared to the short range of alpha particles, could improve the characterization of the RBE.

Moreover, the miniaturization and easy-stable operational use of

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diamond detectors make them suitable for the inclusion in catheters with the purpose of monitoring the dose released to reference points during either external radiotherapy treatments or, more interesting, during brachitherapy applications. High Dose Rate (HDR) brachitherapy is based on the possibility to deliver dose to any target tumor that could be accessed by a catheter or applicator (Zhou et al, 2015). Despite the placement of the catheters is performed under image-guidance, the integration of real-time detectors could provide in-vivo verification of the delivered dose thus to benefit the outcome of the treatment.

Several optimizations on the diamond detector were lately achieved. In particular, the transversal definition of diamond sensitive volume has been improved and the welding material was displaced from the part which defines the sensitive volume of the detector. Otherwise, the welding material would behave as an absorber degrading the energy of ions which cross the sensitive volume and consequently distorting the energy deposition spectra, depending on the particle type and energy (Davis et al., 2014b; Rollet et al., 2012).

In this paper, a novel design of diamond based microdosimeter is presented and all the regions of the detector have been studied in terms of the possible contributions to the charge collection signal. To this purpose, the detector has been characterized by using Ion Beam Induced Charge (IBIC) technique, available at the Ruđer Bošković Institute (RBI) ion microprobe facility. The diamond prototype has been tested with different ions and energies, which are significant in particular for ion beam therapy applications. Finally, the experimental results have been also analyzed in terms of Monte Carlo simulations.

2. Materials and methods

2.1. Diamond based microdosimeter

The diamond based microdosimeter (DBM from now) has a multilayered structure obtained by a two-step growing procedure by Microwave Plasma Enhanced CVD (MWPECVD) technique. Selective CVD deposition of diamond has been adopted in order to accurately define the detector geometry.

The fabrication process consists on the following steps. First, a chromium layer of 300 nm in thickness is thermally evaporated on a commercial, low cost, high pressure high temperature (HPHT) single crystal diamond substrate. A first photolithography step is then performed in order to define the geometry of the back electrode (Fig. 1-a). The p-type diamond is selectively grown by MWPECVD on diamond substrate by using a patterned chromium plasma-resistant mask (Fig. 1-b). A sheet resistance of about 1.5 kΩ/sq was measured by Hall effect measurements at room temperature in air. The p-type diamond electrode has a square shape of 300 × 300 μm^2 in size connected by a 20 μm p-type diamond strip to a second p-type diamond bond-pad (120 μm × 120 μm in size). The chromium mask is then removed by wet chemical etching and an intrinsic diamond layer is selectively homoepitaxially grown by following the same procedure reported above (Fig. 1-c and d). The thickness of the intrinsic diamond layer is about 0.8 ± 0.05 μm as measured by Atomic Force Microscope. The intrinsic diamond layer is oxidized, after the growth, by isothermal annealing at 500 °C for 1 h in air, in order to remove the hydrogen surface conductive layer.

A chromium contact (50 nm thick), aligned over the p-type diamond backing contact, is patterned on the CVD intrinsic diamond surface by using a lift-off photo-lithographic technique (Fig. 1-e). The chromium electrode has a square shape of 300 μm × 300 μm in size. Finally, a second chromium layer, which acts as welding pad electrode (120 μm × 120 μm in size), is realized by using a second lift-off photolithographic technique step (Fig. 1-f). The DBM is schematically shown in Fig.1-g and a SEM image of DBM is reported in Fig.1-h.

A 50 μm aluminum wire is then used to micro-bond the welding pad electrodes (both p-type diamond and metallic contact) in order to provide the electrical connection to the readout electronics.

The microdosimeter is in practice a p-i-metal structure with the metallic rectifying contact having a Schottky barrier of about 1.2 eV as deduced from previous studies (Ciancaglioni et al., 2011). In all of the tests reported in the present work, the DBM was always operated at 0 V.

In order to evaluate the device sensitive thickness, capacitance-voltage (C-V) measurements have been performed using an Agilent 4284A LCR meter. The sensitive volume of the DBM can be approximated by a parallel plate capacitor, whose depletion layer thickness W can be derived as follows: $W = \frac{\varepsilon_0 \varepsilon_r A}{C}$, where A is the active device area (300 μm × 300 μm), ε_0 is the dielectric constant of free space, ε_r (5.7) is the relative permittivity of diamond and C is the device capacitance. Fig. 2 shows the C-V curve for DBM device at room temperature and at 1 kHz frequency in the ± 1 V bias range with a bias step of 0.05 V. The C-V was measured by sweeping the bias from 0 V to + 1 V followed by an opposite direction sweeping. There is not significant hysteresis during the biases sweeping. Using the equation reported above, a depletion thickness of about 0.72 μm is extracted from the C-V measurements at zero bias voltage condition. Such a value tends to about 0.8 μm as the bias voltage is increased, which corresponds to the thickness of intrinsic diamond film obtained by the AFM technique.

2.2. Experimental set-up

The DBM has been characterized at the Ruđer Bošković Institute (RBI) ion microprobe facility (Tadić and Jakšic, 2009). The ions were accelerated by a 6 MV Tandem Van de Graaff accelerator. IBIC technique has been employed to characterize the detector response in terms of its charge collection properties, uniformity of the response and confinement of the sensitive volume.

The detector was placed in vacuum at the end of the beam line (with residual pressure below 10^{-5} mbar) perpendicular to the ion beam direction. Different areas of the sample were irradiated by raster scanning the microbeam on its surface. The size of irradiated areas of the detector were varied in the range from 100 μm × 100 μm up to 500 μm × 500 μm.

The calibration of the beam position and IBIC image dimension was made scanning the microbeam over a silicon diode with a copper wire mask with calibrated micrometric mesh positioned on top of it. The uncertainty of the ion position was estimated in the range 1 μm ÷ 3 μm, depending on the set-up of the focusing magnets, and on the species and energy of the ions. The spot size of the focused ion beam was approximately 1 μm.

The DBM was connected to a conventional charge-sensitive electronic chain, consisting of a charge-sensitive preamplifier ORTEC 142A and an ORTEC 570 shaping amplifier. The shaping time of the amplifier was set to 0.5 μs. Pulse height spectra for the incident ions were measured using an analog to digital multichannel analyzer CAMBERRA 8075.

Different ion beams and energies were used i.e. proton, helium, lithium, carbon and oxygen. The ions employed in the experiment are reported in Table 1. The penetration depths of the different ions in diamond vary between 1.8 μm and 21 μm allowing the ions to cross the DBM sensitive region. The LET profiles of the tested ions as a function of penetration depth in diamond have been calculated by Monte Carlo simulations and reported in Fig. 3. Summary data of the interaction of ions within the diamond detectors have been produced by means of the multi-purpose Monte Carlo tool Geant4 (Geant4.9.10, patch 01 version) (Solevi et al., 2015; Agostinelli et al, 2003). The interactions are computed according to the standard electromagnetic physics, and nuclear stopping effect is based on ICRU49 parameterization. The maximum step length within the sensitive region is set to 10 nm, to sample the energy imparted with the desired precision. The Monte Carlo returns the energy imparted to the sensitive layer per incident ion. The LET is then calculated by dividing the mean imparted energy by the thickness of the diamond layer. For each ion type a sufficient number of histories