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Mechanical properties of the X-ray irradiated DLC films containing SiO_x as a constructive element for radiation detectors

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Abstract

Evolution of physical properties and radiation-induced changes of the film composition and chemical bonding structure have been investigated in ion beam deposited and SiO_x-containing DLC films after their exposure to high-energy X-ray photons generated in medical linear accelerator aiming the possible use of these films as protective coatings or passive layers in the construction of radiation detectors. Mechanical properties of the irradiated films were characterized by microhardness measurements and atomic force microscopy and were analyzed in parallel with the optical properties of the investigated samples. Chemical bonding structure of the DLC films was estimated by Raman spectroscopy (RS). Atomic composition of the films was evaluated from the X-ray photoelectron spectroscopy (XPS) measurements. Changes in surface morphology and increased hardness of the investigated samples as compared to the initial samples were observed. Comparing the results of investigation of SiO_x-containing DLC films with those obtained for undoped hydrogenated DLC films, it was possible to figure out the relationship between radiation-induced structural changes and modification of the properties of different DLC films during irradiation by high-energy X-ray photons.

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Keywords: X-ray photons; SiO_x-containing DLC films; Radiation effects; Bonding structure; Chemical composition; Mechanical properties

1. Introduction

Polycrystalline diamond films are widely used as an active material of the radiation detectors. Diamond-based radiation detectors are very suitable for medical applications, due to the diamond equivalency to the soft tissue ($Z_{\text{eff, diamond}} \approx 6.4$; $Z_{\text{eff, tissue}} \approx 7.1$) as well as their electrical and mechanical properties. Properties of polycrystalline diamond and their behavior in different radiation fields are well investigated [1–3]. Another type of carbon—diamond-like carbon (DLC)—cannot be applied as the active elements of the detectors. However, DLC films are widely used as protective coatings in different applications such as protection of the surface of organic scintillation detectors [4]. It is much easier to synthesize DLC films in comparison with polycrystalline diamond. Due to the possibility of the deposition at room temperature, there are many more substrates suitable for DLC synthesis. Amorphous DLC films are tissue equivalent; they have a smooth surface and are defined by outstanding optical, mechanical and electrical properties [5]. These properties could be improved by doping of DLC films with SiO_x. SiO_x-containing DLC films are defined through reduced internal stress, better adhesion, increased optical transparency and higher thermal stability [6–9]. Combination of optical, electrical and mechanical properties of SiO_x-containing DLC coatings would be of benefit developing protective coatings for Si-based radiation detectors or applying low dielectric constant insulating DLC films as passivating layers in the construction of electronic devices, which are used for radiation detection. However, there is a lack of information about radiation-induced changes in amorphous DLC after their exposure to high-energy X-ray photons.

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The aim of this work was to investigate radiation-induced structural changes and mechanical properties of SiO_x-containing amorphous DLC films produced at room temperature using direct ion beam method [5] after their exposure to high-energy (medical range) X-ray photons with the scope to assess the possibility of their application as protective coatings for radiation detectors.

2. Experimental

Different types of amorphous diamond-like hydrogenated carbon films synthesized at room temperature by direct ion beam method were used in this investigation: 3E/3R samples—amorphous SiO_x-doped DLC films, deposited onto Si⟨1 1 1⟩ wafers from hexamethyldisiloxane and hydrogen gas mixture ($(CH_3)_3SiOSi(CH_3)_3 + H_2$); 4E/4R samples—amorphous SiO_x-containing DLC films co-doped with 2.5% of nitrogen (N_2). Co-doping with N_2 was performed with the purpose to enhance optical transmittance of the films. For the comparison, the properties of amorphous hydrogenated undoped DLC films, synthesized on quartz glass wafers from acetylene (C_2H_2) gas (5E/5R samples), were investigated additionally. All samples were deposited at ion beam energy of 800 eV and ion beam current density of 100 μA cm^{-2} using a closed drift ion source. Working pressure of 2×10^{-2} Pa was kept during the whole deposition process. Thickness of all prepared samples was ~200 nm. Deposition method and some properties of the initial samples have been discussed elsewhere [5,7,8].

The synthesized DLC films were irradiated by high-energy X-ray photons generated at the X-ray tube voltage of 15 MV in medical linear accelerator CLINAC (VARIAN). Total dose of 2 Gy was delivered to each DLC film. Average photon energy of 10.8 MeV was estimated from Monte Carlo calculations [10,11]. Initial samples were indexed by "E" (3E, 4E, 5E) and the irradiated samples had the index "R" (3R, 4R, 5R)

The static microhardness measurements of the samples were performed using Vicker's diamond pyramid indenter [12]. Surface morphology was characterized by atomic force microscope NANOTOP-206. V-shaped ULTRASHARP Si cantilever (force constant 1.5 Nm^{-1}) has been used. The measurements were performed using tapping mode.

Raman spectrometer operating at 514 nm has been used for the investigation of ionization-induced changes in the bonding structure of the irradiated DLC films in the Raman shift range from 900 to 2100 cm^{-1} with 10 cm^{-1} spectral resolution. FTIR spectra were obtained using Spectrum GX FT-IR (PERKIN ELMER) spectrometer. FTIR transmittance and reflectance spectra were detected in the range of 1400–4000 cm^{-1} at a spectral resolution of 0.3 cm^{-1}.

Surface composition of the HMDSO + Si films was analyzed with the X-ray photoelectron spectroscopy (XPS) method. The KRATOS ANALYTICAL XSAM800 spectrometer with nonmonochromatized Al Kα radiation ($hv = 1486.6$ eV) was used. Energy scale of the system was calibrated according to Au 4f7/2 and Cu 2p3/2 Ag 3d5/2 peaks position. The O 1s, C 1s and Si 2p spectra were determined at the 20-eV pass energy (0.1-eV energy increment) and the analyzer being in the fixed analyzer transmission (FAT) mode. Relative surface atomic concentrations were calculated from the appropriate peak area with respect to sensitivity factors, using original KRATOS software.

Film thickness and refractive index were estimated using Laser ellipsometer Gaertner 117 operating with a He–Ne laser ($\lambda = 632.8$ nm).

3. Results and discussions

Radiation detection properties of radiation detectors have to remain stable upon device exposure to ionizing radiation. Due to this reason constructive elements of radiation detectors including protective coatings are characterized through specific properties indicating no significant changes after their irradiation.

Characteristics of the investigated DLC films before and after their irradiation with 15-MeV X-ray photons are presented in Table 1. It was found that adhesion properties and stability of the DLC films were not affected significantly after the irradiation. However, small changes of surface morphology and increased hardness were indicated for all investigated samples. Surface morphology of the initial (3E) and irradiated (3G) samples obtained from the AFM measurements is presented in Fig. 1. It should be noted that the surface roughness increases from 0.6 to 4.0 nm corresponding to the radiation-induced

Table 1
Comparison of DLC film properties before (E) and after (R) their irradiation by high-energy X-ray photons

	3E	3R	4E	4R	5E	5R
Substrate	Si⟨1 1 1⟩		Si⟨1 1 1⟩		Quartz glass	
Reagents	$(CH_3)_3SiOSi(CH_3)_3 + H_2$		$(CH_3)_3SiOSi(CH_3)_3 + H_2 + 2.5\%N_2$		C_2H_2	
Refractive index	1.85	1.7	1.7	1.61	2.3	2.2
Optical band gap (eV)	2.4 (3.12[8])	2.8	2.8 (3.5[8])	2.85	0.9	1.1
Density (g/$cm^{3)}$	1.87	1.91	1.92	1.93	1.79	1.98
Hardness (Gpa)	18.8	20,2	19.1	19.8	16.1	22.0
Surface roughness (nm)	0.6	4.0	1.1	5.4	0.4	0.9