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Effect of heavy ion irradiation on optical property of radiation-crosslinked hydroxypropyl cellulose gel containing methacrylate monomers

A. Hiroki [a,*], S. Yamashita [b], A. Kimura [a], N. Nagasawa [a], M. Taguchi [a]

[a] Quantum Beam Science Center, Japan Atomic Energy Agency, 1233 Watanuki, Takasaki, Gunma 370-1292, Japan
[b] Nuclear Professional School, School of Engineering, The University of Tokyo, 2-22 Shirakata-shirane, Tokai-mura, Naka-gun, Ibaraki 319-1188, Japan

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ABSTRACT

Effects of dose rate and liner energy transfer (LET) on the optical property of a polymer gel dosimeter irradiated with swift heavy ions were investigated. The polymer gel dosimeters that consist of 2-hydroxyethyl methacrylate, polyethylene glycol dimethacrylate, and tetrakis(hydroxymethyl)phosphonium chloride with radiation-crosslinked hydroxypropyl cellulose gel matrix were prepared. The dosimeters were irradiated with 150 MeV/u He ions, 290 MeV/u C ions, and 500 MeV/u Fe ions at HIMAC, and then were optically analyzed by using a UV–Vis spectrophotometer. Absorbance of the irradiated dosimeters increased with an increase in the dose up to 10 Gy. The absorbance at the dose of 5 Gy decreased with increasing dose rate in all of the heavy ions. The dosimeter irradiated with Fe ions exhibited the lowest dose response of the absorbance. It was found that the sensitivity of the dosimeters decreased with increasing dose rate as well as LET of the incident heavy ions.

1. Introduction

In recent years, the radiation therapy techniques have progressed remarkably. In the advanced therapy such as heavy ion radiotherapy and intensity modulated radiotherapy, effective doses are delivered to a cancer while preserving a surrounding normal tissue. The heavy ion radiotherapy using C ions in particular is drawing much attention because of the radiobiological advantage. In addition, heavy ion beams exhibit the increment in linear energy transfer (LET) in a Bragg peak as compared to the entrance region, resulting in a characteristic dose distribution. The three-dimensional dose distribution is calculated for the heavy ion therapy by using a radiation planning system. Also, it is necessary to perform experimental measurement.

Gel dosimeters based on tissue-equivalent gels are drawing attention as a tool of measuring dose distribution. One of the gel dosimeters is Fricke gel dosimeter that consists of the ferrous sulfate (Fricke) solution with either agarose or gelatin gel. Fricke gel dosimeters have been reported for over 30 years, since the three dimensional radiation dosimetry by using Fricke gel dosimeter and magnetic resonance imaging system was proposed [1,2]. Polymer gel dosimeters also have been expected as a useful tool for measuring a three-dimensional dose distribution [3,4]. As the polymer gel dosimeters consisting of some monomers with gel matrix are exposed to radiation, the polymers are formed by the polymerization of the monomers, and then aggregate to produce small particles in the gel matrix, resulting in a light scattering and cloudiness. Conventional polymer gel dosimeters are prepared with toxic monomers and gelatin, which have a risk of the disappearance of the three-dimensional dose distribution because of the re-melting of the gelatin at the temperature over 35 °C. To solve these disadvantages, new polymer gel dosimeters that consist of less toxic methacrylate-type monomers and thermally stable hydrogel have been reported [5,6].

Recently, we proposed a polymer gel dosimeter containing of two methacrylate monomers (2-hydroxyethyl methacrylate and polyethylene glycol dimethacrylate) and an antioxidant (tetrakis(hydroxymethyl)phosphonium chloride) with radiation-crosslinked hydroxypropyl cellulose gel [7,8]. The sheet-type polymer gel dosimeter became cloudy with γ-irradiation at only 1 Gy, and showed the increment in a degree of cloudiness with the dose up to 10 Gy. Dose sensitivity of the dosimeter was adjusted by changing the composition ratio between HEMA and 9G, which increased with the 9G concentration [9]. However, there is no report about the dose response of the dosimeters with heavy ion beam irradiation.

* Corresponding author.

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In this work, effects of the dose rate and LET on the optical property of the dosimeters irradiated with He, C, and Fe ions were investigated to obtain preliminary information toward application on the heavy ion radiotherapy.

2. Experimental

2.1. Materials

Hydroxypropyl cellulose (HPC), which has viscosity of 1000–5000 cP at 2% of aqueous solution, and 2-hydroxyethyl methacrylate (HEMA) were purchased from Wako Pure Chemical Industries, Ltd., Japan. Polyethylene glycol #400 dimethacrylate (9G) was supplied by Shin-Nakamura Chemical Co. Ltd., Japan. Tetrakis(hydroxymethyl)phosphonium chloride (THPC) was obtained from Tokyo Chemical Industry Co. Ltd., Japan. All of the chemicals were used without further purification.

2.2. Preparation of polymer gel dosimeter

HPC aqueous solution of 20 wt%, which is a paste-like state, was put between PET films, and then pressed into a sheet of 1 mm thickness ($150 \times 150\ mm^2$). The sample was sealed in polyethylene/nylon bags after degassing using vacuum apparatus and was irradiated to a dose of 10 kGy with electron beam (2 MV, 2 mA) at Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency (JAEA). The radiation-crosslinked HPC gel was cut into small pieces of $20 \times 30\ mm^2$. The pieces were immersed into an excess amount of distilled water to remove uncrosslinked HPC. The washed HPC gels were vacuum-dried at 40 °C. The dried HPC gels were immersed into aqueous monomer solution consisting of 2 wt% HEMA, 3 wt% 9G, and 0.16 wt% THPC. The swollen gel was taken out from the solution, and immediately vacuum-packed into polyethylene/nylon package that has a thickness of 0.15 mm to obtain sheet-type polymer gel dosimeters. The thickness of thus obtained dosimeters including the package was 1.70 ± 0.10 mm.

2.3. Irradiations

Swift heavy ion irradiation was carried out using Heavy Ion Medical Accelerator in Chiba (HIMAC) at National Institute of Radiological Sciences. Beams of $^{4}He^{2+}$, $^{12}C^{6+}$, and $^{56}Fe^{26+}$ with the respective energies of 150, 290, and 500 MeV/u were provided from the Biological irradiation port in the HIMAC. These ion beams were spread laterally using a pair of wobbler magnets and a scatterer. The circular irradiation region was a diameter of 100 mm, in which the variation in the beam intensity is estimated as 5% or less. The maximum beam intensities of He, C, and Fe ions were 1.2×10^{10}, 2.0×10^{9}, and 2.5×10^{8} ions per second on average, respectively. The polymer gel dosimeters were put to various depths in an acrylic vessel containing water taking into a consideration of quality assurance and quality control applications on the heavy ion radiotherapy, and irradiated horizontally with heavy ion beams, as shown in Fig. 1. Temperature inside the irradiation room was about 24 °C and was controlled with an air conditioner. The LET values at the entrance positions of water are 2.2, 13, and 182 eV/nm, respectively. These values were calculated with Stopping and Range of Ions in Matter (SRIM) code, as previously reported [10]. Dose rates, which were adjusted by using attenuators, were in the range of 0.18–4.31 Gy/min for He ions, 0.09–7.16 Gy/min for C ions, and 0.20–11.62 Gy/min for Fe ions. The dosimeters on the first incidence of ion beams were irradiated at the doses of 1, 2, 3, 5, and 10 Gy. The doses were controlled by the dose monitor located in the upper beam line.

The dosimeters were also irradiated with γ-rays from ^{60}Co source at the Takasaki Advanced Radiation Research Institute, JAEA. Dose rate was 0.17 Gy/min. Temperature inside the irradiation room was about 20 °C.

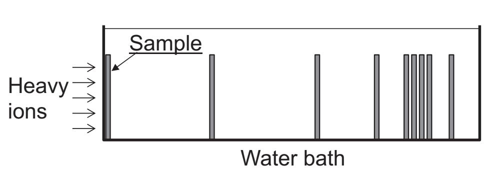

Fig. 1. Illustration of the heavy-ion irradiation to the polymer gel dosimeter. The dosimeters were put to various depths in water bath.

2.4. Optical analysis of irradiated dosimeters

The polymer gel dosimeters irradiated with He, C, and Fe ions were optically analyzed by a UV–Vis spectrophotometer (U-3310, Hitachi High-Technologies Corporation). Optical path was 1.70 ± 0.10 mm because of direct measurement of the dosimeters without using a quartz cell. The dose response of the polymer gel dosimeter was evaluated from the absorbance at 660 nm as a function of the dose. The variation in the absorbance of the dosimeters was in the range of ±30% because of the errors based on the reproducibility of the dosimeter preparation, the thickness of the dosimeters, and the beam homogeneity, as stated above.

3. Results and discussion

Polymerization reactions of the monomers are induced by free radicals such as hydroxyl (OH) radical and hydrated electron produced by water radiolysis [8]. Radiation chemical yield, *G* value, of OH radical and hydrated electron as polymerization initiators depends on a radiation quality. The sum of *G* values of OH radical and hydrated electron was 5.6, 5.0, 4.0, and 2.2 $(100\ eV)^{-1}$ with γ-rays, He ions, C ions, and Fe ions, respectively [11]. Fig. 2 shows the absorbance of the polymer gel dosimeter irradiated with He, C, and Fe ions as a function of the dose. The absorbance of the polymer gel dosimeters at 660 nm increased with an increase in the dose up to 10 Gy, in which dose rates of He, C, and Fe ions were not much different (2.60, 2.13, and 3.28 Gy/min, respectively). The absorbance of the γ-irradiated dosimeter reached 0.319 at 5 Gy, in which the dose rate of γ-rays was 0.17 Gy/min (not shown in Fig. 2). The increment in the absorbance is due to the radiation-induced polymerization of HEMA and 9G. The formed HEMA-9G copolymers aggregated in the HPC gel, resulting in the cloudiness of the dosimeter. The dosimeters irradiated with He, C, and Fe ions exhibited a lower absorbance than dosimeters irradiated with γ-rays. Dose sensitivity is about 0.005 Abs. Gy^{-1} with C ions. This value is far lower than under γ-rays. As mentioned above, *G* value of

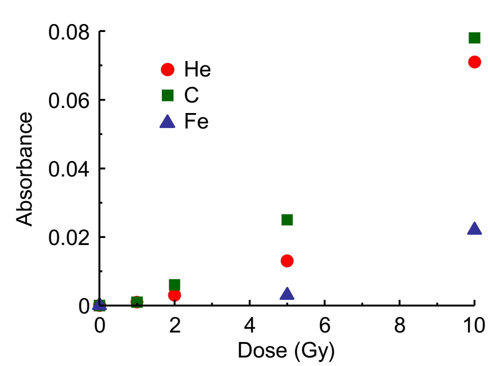

Fig. 2. Dose response of the polymer gel dosimeter irradiated with He, C, and Fe ions.