Enhancement in dose sensitivity of polymer gel dosimeters composed of radiation-crosslinked gel matrix and less toxic monomers

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Abstract. Polymer gel dosimeters based on radiation-crosslinked hydroxypropyl cellulose gel were prepared, which comprised 2-hydroxyethyl methacrylate (HEMA) and polyethylene glycol #400 dimethacrylate (9G) as less toxic monomers and tetrakis (hydroxymethyl) phosphonium chloride (THPC) as an antioxidant. The dosimeters exposed to ^{60}Co γ-rays became cloudy at only 1 Gy. The irradiated dosimeters were optically analyzed by using a UV-vis spectrophotometer to evaluate dose response. Absorbance of the dosimeters linearly increased in the dose range from 0 to 10 Gy, in which dose sensitivity increased with increasing 9G concentration. The dose sensitivity of the dosimeters with 2 wt% HEMA and 3 wt% 9G was also enhanced by increment in THPC.

1. Introduction

Most of the polymer gel dosimeters [1] utilize gelatin and either methacrylic acid or acrylamide [2, 3]. Since both monomers are considered harmful, new recipes for polymer gel dosimeters using less toxic monomer, *N*-isopropyl acrylamide (NIPAM), have been reported [4]. Furthermore, NIPAM polymer gel dosimeters with improved dose sensitivity have been reported by Chain *et al* [5]. Recently, Hayashi *et al* reported that enhancements of dose sensitivity were achieved by adding inorganic salts [6]. On the other hand, Hiroki *et al* reported that the dose sensitivity of dosimeters based on gellan gum and less toxic monomers increased with the initial monomer concentration [7]. In these gel dosimeters, the solubility and compatibility of the chemicals become a hindrance to the increasing dose sensitivity as well as the preparation of transparent polymer gel dosimeters.

Gels of polysaccharide derivatives such as carboxymethyl cellulose and hydroxypropyl cellulose (HPC) were prepared by γ-rays or electron beam irradiation to their highly concentrated aqueous solution without any crosslinker. The prepared gels have been studied to apply to a soil improvement material in agriculture and a radiation dosimeter in radiation therapy, using a high water-absorption [8-10]. It is possible to prepare the polymer gel dosimeter with a high monomer concentration, since the gel largely absorbs not only water, but also organic solvent.

This study aims to enhance in dose sensitivity of polymer gel dosimeters based on a radiation-crosslinked gel matrix. HPC gel was selected as gel matrix for the dosimeter because of transparent and thermostable material in comparison with gelatin gel. Polymer gel dosimeters comprising less toxic methacrylate monomers, THPC, and HPC gel matrix were prepared, and then exposed to γ-rays.

Dose response was evaluated by measuring absorbance of the dosimeters. Effects of the composition ratio of monomers and THPC concentration on the dose sensitivity of the dosimeters were investigated.

2. Materials and methods

2.1. Preparation of polymer gel dosimeters

HPC was dissolved in ultrapure water at the concentration of 20 wt%. The HPC aqueous solution as a paste-state was formed to 1.0 mm thickness by pressing, and then was irradiated to a dose of 10 kGy with electron beam (2 MeV, 2 mA) at the Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency. The obtained HPC gel membrane was immersed into an excess amount of distilled water to remove uncrosslinked HPC, and then vacuum-dried. The dried HPC gels were immersed into monomer aqueous solutions that consist of 0.8-4 wt% HEMA, 1-6 wt% 9G, and 0-0.24 wt% THPC. The swollen HPC gels were vacuum packed in PE/nylon package to obtain polymer gel dosimeters. The prepared samples were stored in a refrigerator until γ-irradiation.

2.2. Irradiation

The prepared samples were irradiated up to 10 Gy by using a ^{60}Co g-ray source at the Takasaki Advanced Radiation Research Institute, Japan Atomic Energy Agency. Dose rate was adjusted by varying the distance between the samples and the source, which was in the range of 3.0 to 30 Gy h^{-1}. The temperature inside the irradiation room was about 20ºC and was controlled with an air conditioner. After irradiation, all samples were stored in the refrigerator.

2.3. Analysis of irradiated samples

The absorbance of the polymer gel dosimeters were measured after 24 hours post-irradiation. The optical analysis was carried out using an UV-vis spectrophotometer (Hitachi High-Technologies Corporation, U-3310) at 25ºC. Dose sensitivity of the dosimeter was estimated from the absorbance at 660 nm as a function of the dose.

3. Results and Discussion

Figure 1 shows photographs of non- and γ-irradiated polymer gel dosimeters. They became cloudy by g-irradiation at 1 Gy. It is clearly seen that the cloudiness of the polymer gel dosimeters prepared by using a monomer solution with 2 wt% HEMA, 3 wt% 9G, and 0.16 wt% THPC increased with the increasing dose.

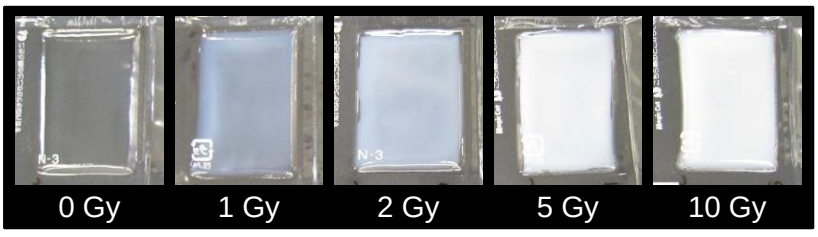

Figure 1. Photographs of non- and γ-irradiated polymer gel dosimeters consisting of HEMA, 9G, and THPC with HPC gel. HEMA, 9G, and THPC concentrations of the monomer solution were 2 wt%, 3 wt%, and 0.16 wt% respectively.

Figure 2 shows the absorbance of the polymer gel dosimeters with various composition ratios between HEMA and 9G as a function of dose. The absorbance of the polymer gel dosimeters increased approximately linearly in the dose range from 0 to 10 Gy. Dose sensitivity defined as the initial increment in absorbance per unit dose increased with an increase in 9G concentration, reached

about 0.04 Abs Gy^{-1} at the dosimeter with 2 wt% HEMA, 3 wt% 9G. Maryanski *et al* also reported that the dose sensitivity of BANG polymer dosimeter significantly depended on the composition ratio between acrylamide (AAm) and *N,N'*-methylene bis-acrylamide (MBAAm) as a crosslinker [11]. A crosslinker induces an enhancement of gel fraction because it promotes polymerization [12]. 9G as a crosslinker would promote the formation of a copolymer gel comprised of HEMA and 9G. Small particles are produced with irradiation in 9G-rich condition in comparison with HEMA-rich one [10]. It is considerable, therefore, that the polymer conversion and the aggregation size enhance the increase in the absorbance, resulting in the increase in the dose sensitivity.

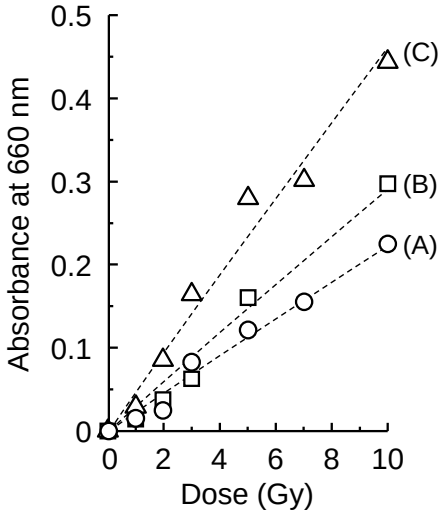

Figure 2. Increment of the absorbance of the polymer gel dosimeters prepared with aqueous solutions consisting of HEMA, 9G, and THPC. HEMA and 9G concentrations are 4 wt% and 1 wt% (A), 3 wt% and 2 wt% (B), and 2 wt% and 3 wt% (C), respectively. THPC concentration is the same in 0.16 wt%.

To induce higher absorbance in the polymer gel dosimeter, it is preferable to increase in polymerization product scattering light. The polymer gel dosimeters with various total monomer concentrations (2, 5, and 10 wt%) at composition ratio of 2/3 were prepared. As a result of the irradiation at 10 Gy, the absorbance of the dosimeters increased with total monomer concentration, reaching about 1.3 at 4 wt% HEMA and 6 wt% 9G.

The polymerization reaction is inhibited in oxygen-contaminated gel, since oxygen scavenges free radicals produced by water radiolysis. THPC as an antioxidant plays an important role for reduction of the oxygen effects. The absorbance of the dosimeters with various concentrations of THPC increased in the dose range from 0 to 10 Gy. Figure 3 shows relationship between THPC concentration and the dose sensitivity of the polymer gel dosimeters with 2 wt% HEMA and 3 wt% 9G. The dose sensitivity increased with the initial THPC concentration. Jirasek *et al* reported that optimum THPC concentration to maximize the dose response in the PAGAT dosimeter is about 4.5 mM because of the reactions with gelatin, AAm, and MBAAm as gel constituents at high concentration [13]. This is corresponded to about 0.09 wt% THPC. In the dosimeter consisting of HEMA and 9G with HPC gel, there is no maximum of the dose sensitivity until 0.24 wt% THPC. This would be due to the effects of the matrix and monomers.

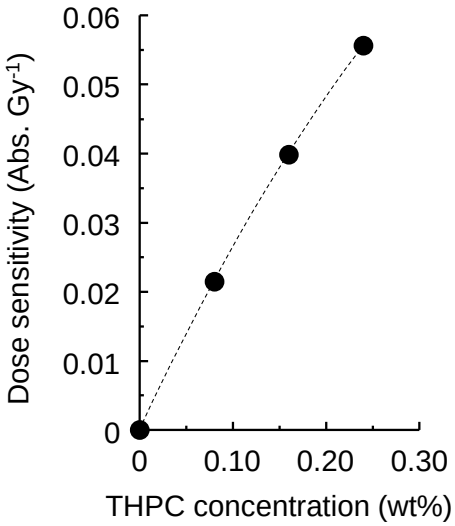

Figure 3. Relationship between THPC concentration and dose sensitivity of the prepared polymer gel dosimeters with 2 wt% HEMA and 3 wt% 9G.

4. Conclusion

Polymer gel dosimeters that consist of HEMA, 9G, and THPC with radiation-crosslinked HPC gel showed the cloudiness at 1 Gy to be observable easily by eyes. The absorbance of the polymer gel dosimeter increased linearly in the range of dose from 0 to 10 Gy. The dose sensitivity increased with 9G concentration. The dose sensitivity could be also enhanced by adjusting the THPC concentration, reached about 0.056 Abs. Gy^{-1} at the dosimeter with 2 wt% HEMA, 3 wt% 9G, and 0.24 wt% THPC.

5. Acknowledgement

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6. References

[1] Baldock C *et al* 2010 *Phys. Med. Biol.* **55** R1-63
[2] Maryanski M J *et al* 1994 *Phys. Med. Biol.* **39** 1437-55
[3] Baldock C *et al* 1998 *Phys. Med. Biol.* **43** 695-702
[4] Senden R J *et al* 2006 *Phys. Med. Biol.* **51** 3301-14
[5] Chain J N M *et al* 2011 *Phys. Med. Biol.* **56** 2091-102
[6] Hayashi S *et al* 2012 **81** 884-8
[7] Hiroki A *et al* 2013 *Phys. Med. Biol.* **58** 7131-41
[8] Hiroki A *et al* 2011 *Trans. Mater. Res. Soc. Jpn.* **36** 397-400
[9] Hiroki A *et al* 2013 *J. Phys.: Conf. Ser.* **444** 012028
[10] Yamashita S *et al* 2014 *Radiat. Phys. Chem.* **101** 53-8
[11] Maryanski M J *et al* 1997 *Phys. Med. Biol.* **42** 303-11
[12] Sajjadi S *et al* 1996 *Polymer* **37** 4141-8
[13] Jirasek A *et al* 2006 *Phys. Med. Biol.* **51** 1891-906