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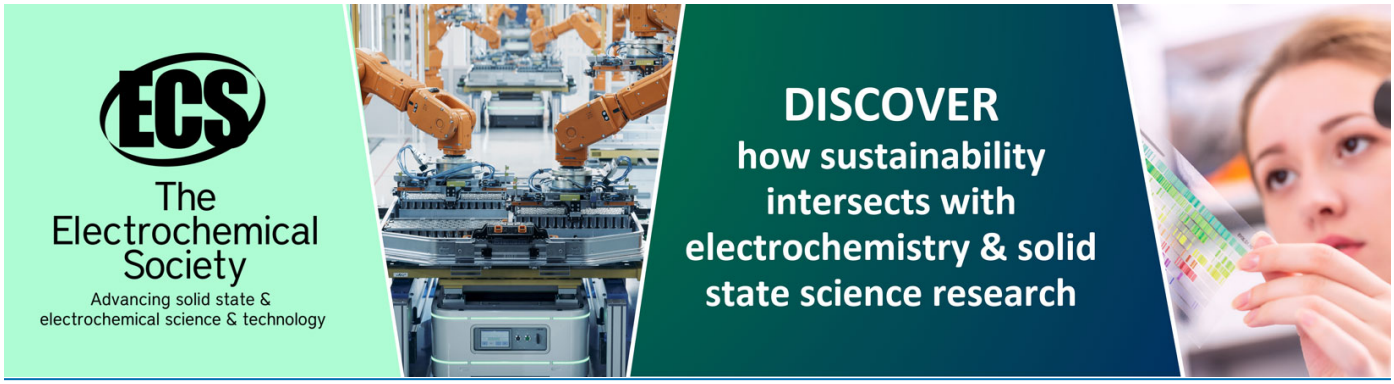

Effects of PVA-GTA-I radiochromic gel dosimeter components on optical dose-response

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Abstract. In recent years, a novel radiochromic gel dosimeter was developed that utilizes the color development of a polyvinyl alcohol-iodide (PVA-I) complex. In this study, we explore the effects of different iodide salts (LiI, NaI, KI, CsI, NH_4I, CaI_2, and ZnI_2) and PVAs with different degrees of polymerization (500, 1000, and 1500) and saponification (80, 88, and 98 mol%) were investigated on a PVA-GTA-I gel dosimeter using PVA that was chemically crosslinked with glutaraldehyde (GTA) as a matrix. The results showed that these substitutions had negligible effect on dose-responses, such as sensitivity and dose-rate independence.

1. Introduction

Recently, we developed a novel radiochromic PVA-I gel dosimeter [1, 2]. The dosimeter utilizes the red color development due to the complexation of polyvinyl alcohol (PVA) with iodide (I_3^-) formed by irradiation in an aqueous solution. The PVA-I gel is composed of partially saponified PVA, potassium iodide (KI), fructose as a reductant, gellan gum as a gelling agent, and distilled water. This gel dosimeter has excellent dose characteristics such as high sensitivity, dose rate independence, and a wide dose range. In particular, the PVA-I gel dosimeter can be decolored (initialized) by heating, owing to the reducing nature of fructose, resulting in its reusability. In this gel dosimeter, PVA acts as a host for iodide ions to form a complex. Therefore, a gelling agent is required for the gelation of the solution. The gellan gum used in the PVA-I gel dosimeter is solidified by physical cross-linking with cations.

On the other hand, a Fricke-type gel dosimeter using PVA chemically crosslinked with glutaraldehyde (GTA) as a matrix was reported and named PVA-GTA Fricke gel dosimeter [3, 4]. Employing the PVA-GTA matrix in the Fricke gel dosimeter resulted in high transparency, low diffusivity of ferric ions created by irradiation, and higher dose sensitivity. If the PVA-GTA matrix can be applied to the PVA-I gel dosimeter, then no gelling agent would be required, resulting in higher transparency. Therefore, a PVA-I gel dosimeter using a PVA-GTA matrix (PVA-GTA-I gel dosimeter) was developed [5, 6].

In the present study, we investigated the effects of different iodides (LiI, NaI, KI, CsI, NH_4I, CaI_2, and ZnI_2), the average degree of polymerization (DP: 500, 1000, and 1500), and the degree of saponification (DS: 80, 88, and 98 mol%) of PVA on the dose response of the PVA-GTA-I gel dosimeter.

Table 1 Components of the PVA-GTA-I gel dosimeter prepared in this study. The standard composition is marked in bold. M is molarity (mol/1000gH_2O).

Base solution									
PVA	Average degree of polymerization			**500**	1000	1500			10 wt%
	Degree of saponification (mol%)		80	**88**	98				
Water									90 wt%
Additives									
Iodide		LiI	NaI	**KI**	CsI	NH_4I			100 mM
							CaI_2	ZnI_2	50 mM
Reducing sugar				**fructose**					100 mM
Cross-linker				**GTA**					10 mM
Proton generator				**GDL**					100 mM

2. Materials and methods

2.1 Gel preparation

The composition of the PVA-GTA-I gel dosimeters manufactured in this study is listed in Table 1. Iodide salt, fructose, GTA, and glucono-δ-lactone (GDL) were added to a 10 wt% PVA solution prepared in advance. GDL was employed as a proton generator to catalyze crosslinking. All the reagents were completely dissolved using a magnetic stirrer at room temperature (15-20 °C). The obtained mixed solution was divided into PMMA cuvettes (4.5 mL, 1 cm path length). Before irradiation, the samples were initialized and solidified by annealing at 45 °C for 12 h in an incubator.

2.2 Irradiation

Similar to a previous study [2], all samples were irradiated using a 6 MV X-ray from a medical linear accelerator (Varian/BrainLAB Novalis Tx, USA) at a depth of 5 cm in solid water phantom at room temperature (20-25 °C). Doses of up to 10 Gy were delivered to the samples, with an average dose rate fixed at 600 cGy/min. Different dose rates of 100, 200, and 400 cGy/min were used to investigate the dose rate dependence.

2.3 Measurement

The absorption spectra of the samples were also measured as described in a previous study [2], one day after irradiation, using a UV-Vis spectrophotometer (UV-1600PC, Shimadzu, Japan). Absorbance (Abs) was calibrated using distilled water. The change in absorbance (ΔAbs) was calculated by subtracting the absorbance of the non-irradiated sample from that of the irradiated sample.

3. Results and Discussion

3.1 Absorption spectra

Figure 1(a) shows the PVA-GTA-I gel dosimeters irradiated with up to 10 Gy. As the absorbed dose increased, their appearance gradually changed from colorless to red, similar to that observed for the PVA-I gel dosimeters [1, 2]. Fig. 1(b) shows the absorption spectra of PVA-GTA-I gel dosimeters. Single peaks centered at approximately 486 nm were exclusively observed. Therefore, the absorbance at 486 nm was used in subsequent investigations to determine the dose responses. However, as shown in Fig.1(c), the spectra of the PVA-GTA-I gel containing PVA with a DS of 98 mol% were different from the others. It displays a remarkable absorbance at 358 nm due to the free iodide ion (I_3^-) that did not form a complex with PVA, while only a small absorption increase was observed at 486 nm. In addition, Fig.1(d) shows the background absorption spectra of PVA-GTA-I and PVA-I gel dosimeters before irradiation (immediately after initialization). The background absorbance (Abs = 0.002) of PVA-GTA-I at 486 nm was substantially lower than that of PVA-I (Abs = 0.040), resulting in higher transparency (lower background).

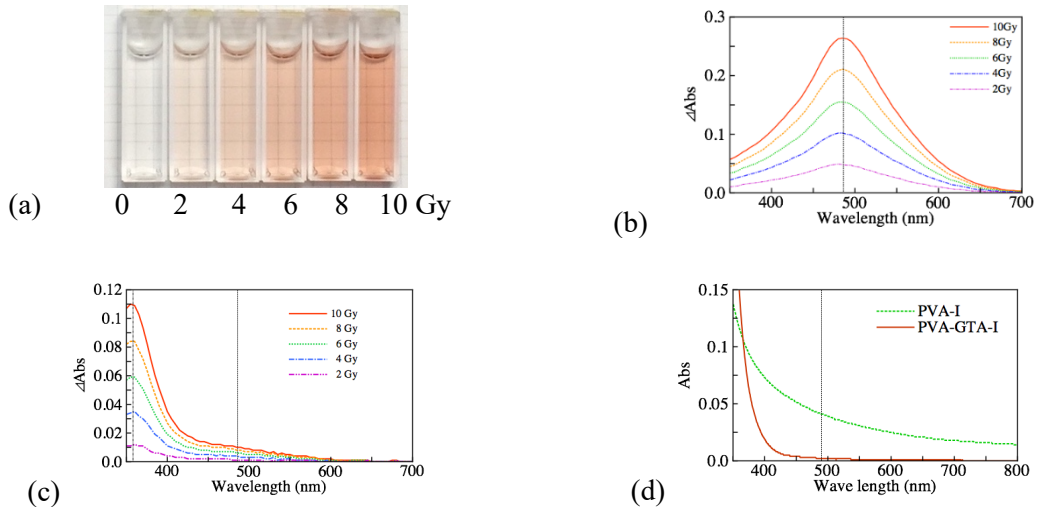

Figure 1 (a) PVA-GTA-(Na)I gel dosimeters irradiated up to 10 Gy. (b) Absorption spectra of PVA-GTA-(Na)I gel dosimeters irradiated up to 10 Gy. (c) Absorption spectra of PVA-GTA-I gel dosimeters containing PVA with 98 mol% DS irradiated up to 10 Gy. (d) Absorption spectra of non-irradiated PVA-GTA- I gel and PVA-I gel dosimeters immediately after initialization (annealing).

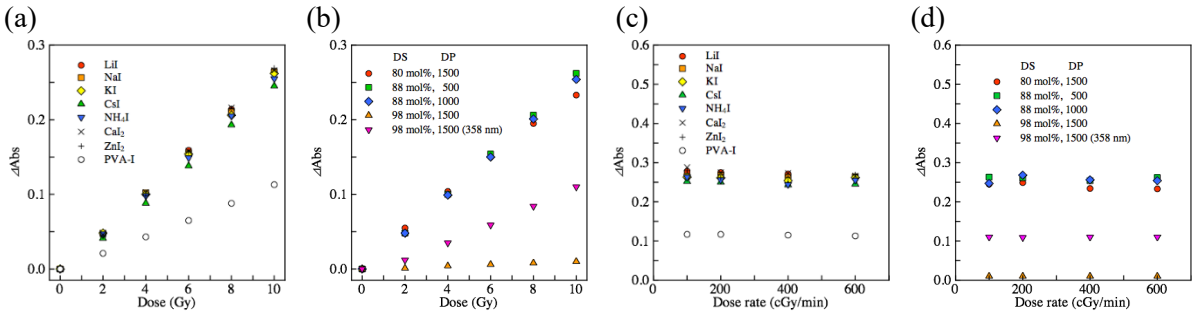

Figure 2 (a) and (b) show the dose responses of the PVA-GTA-I gel dosimeters containing different iodide salts and PVAs with different DSs and DPs, respectively. (c) and (d) represent the dose-rate dependence of the absorbance at the same dose (10 Gy) in the corresponding gel dosimeters. White circles (○) show the response of the PVA-I gel dosimeter for comparison [2]. The data points, except for PVA-I and 98 mol% DS, nearly superimpose each other.

3.2 Dose-responses

3.2.1 Effect of Iodide type

Figure 2 (a) shows the dose response of the PVA-GTA-I gel dosimeters containing different iodide salts. The sensitivities were similar, regardless of the type of iodide salt used. The results indicated that the cation does not affect the dose response of a high-energy photon beam used in radiotherapy, although heavy atoms, such as Cs, could affect the sensitivity in the lower energy regions due to photoelectric absorption. In addition, the sensitivities of the PVA-GTA-I gel dosimeters were approximately 2.5 times higher than those of the PVA-I gel dosimeter [2].

3.2.2 Effect of degree of polymerization (DP) and saponification (DS) of PVA
Figure 2 (b) shows the dose responses of PVA-GTA-I gel dosimeters fabricated with PVAs of different DPs and DSs. The results demonstrated that the sensitivity was only slightly affected by DP and DS, except for that containing PVA with a DS of 98 mol%. This indicates that the number of acetyl residues on PVA is not sufficient to form the maldistributed groups required to form a complex with triiodide ions. The dose response at 358 nm, corresponding to free iodide, showed adequate linearity and sensitivity. This is consistent with a previous study on dosimetry using an aqueous KI solution [7]. However, dosimetry using a PVA-I complex provides higher sensitivity and visibility than dosimetry using a KI aqueous solution. On the other hand, a 10 wt% PVA base solution containing PVA with DS of 80 mol% or a higher DP exhibited higher viscosities than the other PVA aqueous solutions. Therefore, the authors recommend fabricating a gel dosimeter using PVA with lower viscosity, DP of 500–1000, and DS of 88 mol% for the uniformity of the gel.

3.3 Effect of dose rate dependence
Figures 2 (c) and (d) show the absorbance of PVA-GTA-I gel dosimeters containing various components irradiated at 10 Gy at different dose rates. It was shown that dose-rate-independence, which is one of the favorable characteristics of PVA-iodide-based gel dosimeters [1, 2], is also preserved by the substitution of different iodide salts or PVAs in the PVA-GTA-I gel dosimeter.

4. Conclusion
In the present study, the effects of various components on the dose response of the PVA-GTA-I radiochromic gel dosimeter were investigated. The difference in the iodide salts or PVAs had negligible effect on the dose properties of the PVA-GTA-I gel dosimeter, except for the gel dosimeter containing 98 mol% DS of PVA. These results could help optimize the selection of the ingredients for gel dosimeters. Moreover, the PVA-GTA-I gel dosimeter demonstrated higher transparency and sensitivity than those of the PVA-I gel dosimeter. Based on these results, the PVA-GTA-I gel dosimeter exhibits the potential to be a more effective dosimetry tool for radiotherapy. However, further optimization of the composition and evaluation of its fundamental properties, such as temporal, spatial, and thermal stabilities, are still required. Efforts are currently underway to address these issues.

5. Acknowledgment
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6. References
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