

# 論文内容要旨

Effect of the glucono- $\delta$ -lactone concentration on the sensitivity and stability of PVA-GTA-I radiochromic gel dosimeter  
(PVA-GTA-I 色素ゲル線量計の感度と安定性に及ぼすグルコノデルタラクトン濃度の影響)  
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## **Effect of the glucono- $\delta$ -lactone concentration on the sensitivity and stability of PVA-GTA-I radiochromic gel dosimeter**

(PVA-GTA-I 色素ゲル線量計の感度と安定性に及ぼすグルコノデルタラクトン濃度の影響)

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### **Introduction**

The current and emerging techniques in radiation therapy are becoming more complex and the quality assurance of these procedures require more accurate methods to quantify ionizing radiation fields in three dimensions (3D). Conventional dosimetry systems such as ionization chambers and radiosensitive films are utilized in clinical settings to verify dose delivery in the modern radiotherapy. However, these dosimeters do not meet the Resolution-Time-Accuracy-Precision (RTAP) criteria for 3D dosimetry and are limited to providing point or two-dimensional (2D) absorbed dose estimates and sporadic 3D information. Accordingly, such requisite characteristics could be addressed by gel dosimeters which are chemical-based dosimeters capable of retaining spatial dose distribution within the gel matrix. The 3D dose information in the gel dosimeters can be measured by several imaging techniques. The characteristics of gel dosimeters are advantageous in radiotherapy conditions where steep dose gradients are present (e.g., intensity modulated radiation therapy (IMRT) and stereotactic radiosurgery (SRS)). Moreover, the materials used in fabricating most gel dosimeters are radiologically equivalent to soft-tissues, which can be modified depending on the type of application.

Gel dosimeters are categorized depending on their properties and read-out process, these include Fricke gels, polymer gels, and novel radiochromic gels. In more recent years, there have been considerable interest in the development and improvement of radiochromic gel dosimeters due to its potential and relatively easier method of measurement. The mechanism of radiochromic gel dosimeters is based on the radiation-induced color change which is proportional to the absorbed dose. The color change can be measured using optical techniques such as spectroscopy or OCT.

A recently developed radiochromic gel dosimeter composed of polyvinyl alcohol (PVA)-iodide (I) complex with gellan gum (GG) as the gelling agent was reported. This gel dosimeter converts from colorless to red after irradiation and can be reused by heating. The red coloration of this gel results from the oxidation of the PVA-Iodide complex caused by the radiolysis of the water molecules. Whereas the decolorization is attributed to the additive fructose which acts as the reducing sugar. In other studies, the reagent glutaraldehyde (GTA) was utilized as a crosslinker to the PVA matrix of Fricke-XO gel (FXG) dosimeters. PVA is widely used in fabricating gel dosimeters due to its low diffusion coefficient. Additionally, GTA has been proven as an effective crosslinker to PVA and can further reduce the spatial diffusion and increase the sensitivity of gel dosimeters.

We developed our own system radiochromic gel dosimeter, called the PVA-GTA-I gel dosimeter, by applying the PVA-GTA matrix to the PVA-I radiochromic gel dosimeter. Our present research focuses on the effect of one of the additives in our formula, which is the glucono- $\delta$ -lactone (GDL). GDL is an acidic coagulant commonly used in the food industry, such as in bean curd and cheese production. Because of its inherent binding and non-toxic properties, GDL is used in this gel dosimeter formula as a proton generator and to further promote the cross-linking process. Therefore, our objective is to investigate the effect of the GDL concentration to the dose-response and temporal stability of the PVA-GTA-I formula.

### **Materials and Methods**

All gel samples were fabricated with ultrapure water, and analytical grade chemicals. The base solution is made of PVA (86–90 mol% saponification, partially hydrolyzed) that was dissolved in water using a magnetic stirrer at 80°C for 1 h. Then, other components of potassium iodide (KI), fructose, GTA, and GDL were poured into the mixture and stirred until a homogeneous solution was achieved. Six sets of gels were prepared with various GDL

concentrations from 50 to 300 mM. The gel solutions were poured into PMMA cuvettes, covered with polyethylene (PE) cover, and stored in a dry heat sterilizer at 45°C for 12 h to allow gelation. The gel samples were prepared and heated the day before irradiation and were stabilized for approximately 1 h after heating at room temperature before exposure to radiation. A Gammacell-40 research irradiator with low dose rate (i.e., 0.82 Gy/min) <sup>137</sup>Cs sources was used to irradiate the gel samples with doses from 1 to 10 Gy; and one sample was left unirradiated as the control sample. The gel dosimeters were positioned at the middle of the sample holder with the axes of the cuvettes perpendicular to the source. A UV-Vis Spectrophotometer was used to measure the optical absorbance at the wavelength range of 350–800 nm. Ultrapure water was used to calibrate all the absorbance measurements. The change of absorbance ( $\Delta$ Abs.) was then calculated by the difference between the measured absorbances of the irradiated sample and the unirradiated (control) sample. Finally, the temporal stability of the gel was evaluated in a seven-day period after irradiation with time intervals of 2, 24, 48, 72, 96, 120, 144, and 168 h at room temperature range of 20–23°C.

### Results and Discussion

The physical appearance of the unirradiated PVA-GTA-I gel sample was colorless and transparent, while the irradiated samples were converted to red. The color intensity was observed to be increasing with radiation dose. This effect was further emphasized in the absorbance spectrum profiles where the peak absorbance at 482 nm was increasing. The  $\Delta$ Abs. plots at 482 nm peak showed good linearities ( $R^2 < 0.99$ ) in the dose range of 0–10 Gy from all the sample sets despite the different GDL concentrations. The sensitivity, defined by the slope of the linear fitting function, increased with higher GDL concentrations. However, it was also observed that the increment of sensitivity became small with the GDL concentration and then completely saturated around 250 mM. The highest sensitivity range from all the sample sets was  $3.8\text{--}4.0 \times 10^{-2} \text{ Gy}^{-1}$ . These values are approximately 3 to 4 times higher than the reported sensitivity of the PVA-I gel, and 6 to 9 times higher than LCV micelle gels. Though, the sensitivity of the PVA-GTA-I gel was approximately 2 times lower than the PVA-GTA-FXG gels. The absorbance-time plot of the control (0 Gy) samples showed a gradual increase in absorbance with time. This phenomenon is caused by the autooxidation of the iodide ions in the gel. Moreover, it was also seen that higher GDL concentrations promotes the rate of auto-oxidation in the gel formula. The temporal stability results of the 10 Gy irradiated samples revealed the trends of the  $\Delta$ Abs.-time plots for the varying GDL concentrations. The sample sets with 100 to 300 mM GDL exhibited a sharp increase of the  $\Delta$ Abs during the initial 24 h and followed by small fluctuations up to 168 h (7 days) post irradiation. On the other hand, the sample set with 50 mM GDL showed a gradual rate of increase in  $\Delta$ Abs, which started at 48 h after irradiation, and then continuous to increase up to 168 h post-irradiation.

### Conclusion

The influence of the increasing concentrations of GDL to the dose-response and temporal stability of a PVA-GTA-I radiochromic gel dosimeter has been investigated. The PVA-GTA-I gel showed linear dose-response up to 10 Gy with sensitivities 3-9 times higher than other radiochromic gel dosimeters. The temporal stability data revealed that higher GDL concentrations increases the auto-oxidation rate of the PVA-GTA-I gel. Overall, the results obtained in our study have indicated good insights in the properties of the PVA-GTA-I gel dosimeter with respect to varying GDL concentration. Subsequently, the PVA-GTA-I gel dosimeter has shown good potential in reusability from our pilot study, which is an advantageous characteristic for clinical 3D dosimetry applications in radiotherapy. Further research is recommended to uncover more of its characteristics in terms of its reusability after repeated heating and irradiations, dose rate dependence, dose fractionation effects, and spatial stability.