Preparation and characterization of FPGX hydrogel dosimeters

Xiao Ruijia[1] Yang Liming[1,*] Chen Jie[1] Qian Qiang[1] Rong Liang[1] Cao Fangqi[1]
Luo Wenyun[1] Dong Xiaoqing[1] Zha Yuanzi[2] Wu Guohua[2]

[1] *School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China*

[2] *Department of Radiation Oncology, Xin Hua Hospital Affiliated to Shanghai Jiao Tong University School of Medicine, Shanghai 200092, China*

Abstract A three dimensional Fricke-(PVA-glutaraldehyde)-xylenol orange (FPGX) hydrogel dosimeter was prepared by chemical crosslinking method. Dose determination was performed by measuring the absorbance change of the dosimeter before and after irradiation using UV-Vis spectrophotometer at 585 nm. The dose response of the FPGX hydrogel dosimeter related to absorbance change was linear, when it was irradiated by X-ray in the range of 0~2 Gy. The stability of the dosimeter before and after irradiation was evaluated by studying the change of absorbance with time.

Key words Dosimeter, PVA hydrogel, Glutaraldehyde, Crosslinking, Radiotherapy

A growing interest has been focused on the dose measurement for safety and efficiency of the radiotherapy[1]. Ion chambers and film dosimeters are commonly used in measuring the dose of single point or two-dimensional (2D) dose mapping[2], while progresses have been made in developing new types of dosimeters for 3-D dosimetry distributions in complex radiotherapy [3–7]. A three dimensional gel dosimeter system proposed by Duzenli C *et al*[8] in 1994. It was a mixture of Fricke solution and a gelatin (or agarose[9]). Under ionizing radiations, the Fe^{2+} ions in the Fricke gel dosimeter are changed into Fe^{3+} ions to form a colored complex (xylenol orange-Fe^{3+}, or XO-Fe^{3+}) with the xylenol orange [10,11]. Potentially, 3-D dose distributions can be measured as the (XO-Fe^{3+}) is fixed spatially by the gel[12–14]. However, diffusion of ions in the Fricke gel dosimeter is a problem on stability of the dosimeters. Chu *et al.* [15] reported the PVA (polyvinyl alcohol) gel Fricke dosimeter system, in which the PVA gel was solidified via a freeze-thaw process, and the stability of the PVA-gel dosimeter system was better than other gel dosimeter systems reported previously [16].

To improve thermo-stability and transparency of the PVA gel Fricke dosimeter systems, we prepared the FPGX gel dosimeters by chemical crosslinking. Sensitivity and stability of the dosimeters prepared in different conditions (oxygen removal, and storage and measurement temperature) were investigated.

The PVA-AH-26, glutaraldehyde (GA, aqueous solution of 25%), $FeSO_4 \cdot 7H_2O$, H_2SO_4 (98%) and XO (xylenol orange) were of reagent grade, from Sinopharm Chemical Reagent Co Ltd., China, and were used without any purification.

PVA aqueous solution (10wt%) was prepared by dissolving 10 g PVA in 100 mL ultrapure water at 90°C, stirring for 3–5 h and cooled to room temperature. The Fricke dosimeter solution was prepared according to the optimal compositions in Ref.[17]: XO, 0.015 mM; $FeSO_4 \cdot 7H_2O$, 0.1 mM; and H_2SO_4, 25 mM[17]. PVA/GA solution mixture (PVA/GA = 0.01wt%) was prepared by mixing 10% PVA(100 mL) and 25% GA (0.04 mL) solutions under stirring for 5 min at 50°C. The solution was stirred with or without bubbling nitrogen to remove dissolved oxygen, so as to study the oxygen effect. The solution was quickly mixed with the PVA-GA mixture and filled into 4.5-mL cuvettes of 10-mm length. The Frick-(PVA-GA)-XO hydrogel dosimeter was formed in 30 min via the reaction schemed in Fig 1.

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* Corresponding author. *E-mail address:* Yanglm@shu.edu.cn
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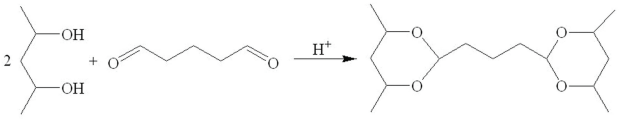

Fig. 1 Reaction scheme of PVA and glutaraldehyde.

X-rays from a 6 MV medical linac (Synergy® IGRT, Elekta, Sweden) was used, at dose rate of 400 MU/min and the source-skin distance (SSD) of 100 cm. The FPGX dosimeter samples were irradiated to 6 Gy, with an increment of 0.5 Gy, respectively.

The irradiated FPGX dosimeters changed color from light-orange to purple with increasing doses. Optical absorbance of the dosimeter at 585 nm, where the absorption peak of XO-Fe^{3+} complex locates, was measured by UV-vis (8453, Agilent, USA).

In Fig. 2, the absorbance of the FPGX dosimeter irradiated to 0–6 Gy changed rapidly with the dose up to 2 Gy, where it began to slow down and saturate. The dose-response is a linear up to 2 Gy, which is a usual fraction of the total dose in radiotherapy. The line has a slope of 0.09917 Gy^{-1}(R^2 = 0.9969), which can be supposed as the dosimetry sensitivity[10]. The experiment was done with 0.1 mM $FeSO_4$, and it showed a better dose response in the lower dose range than our previous work[17]. It might be considered that most of the 0.1 mM Fe^{2+} in the sample had changed into Fe^{3+} at 2 Gy, where the absorbance change tended to a termination.

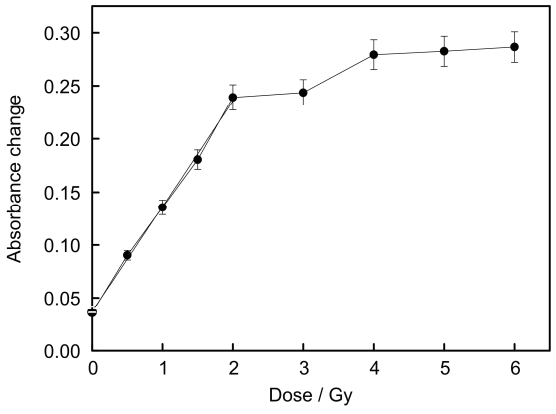

Fig. 2 Absorbance change of irradiated FPGX gel dosimeters.

The changing rate of the absorbance per unit dose ($Gy^{-1}\cdot h^{-1}$) related to time can be expressed as stability of the irradiated dosimeter [10]. This change of the FPGX hydrogel dosimeter was determined as a function of storage time at different temperatures. In Fig.3, slopes of the irradiated FPGX dosimeters, and stored at 5°C or 25°C, were 6.79×10^{-4} $Gy^{-1}\cdot h^{-1}$ (R^2 = 0.9927) for 1 Gy and 8.75×10^{-5} $Gy^{-1}h^{-1}$ (R^2 = 0.9911) for 2Gy. This also indicates that most of the Fe^{2+} ions in the gel dosimeter irradiated to 2 Gy were exhausted.

It was reported that the storage temperature affected stability of the dosimeter [18], and low storage temperature enhanced polymer-polymer network formation, which restricted mobility of the Fe^{2+} and Fe^{3+}[15,16] ions. In Fig.3, however, no obvious difference can be seen between the samples stored at 5°C and 25°C. It may be assumed that the cross-linking of the PVA and glutaraldehyde led to a good thermo-stability of the FPGX hydrogel dosimeter and restricted the motion of ions as well.

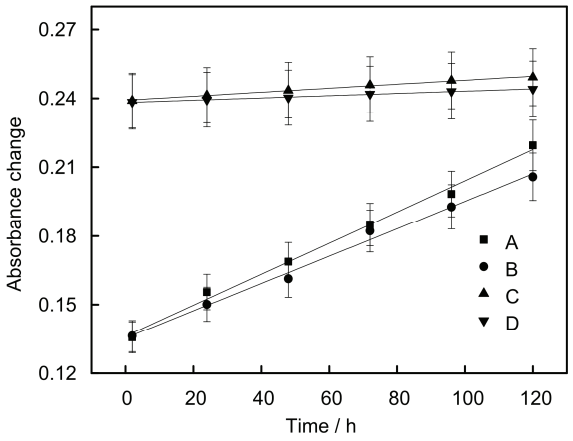

Fig. 3 Effect of storage time on absorbance change of the irradiated FPGX dosimeters stored at different temperatures.
(A) Irradiated to 1 Gy and stored at 25°C (R^2 = 0.9927),
(B) Irradiated to 1 Gy and stored at 5°C (R^2 = 0.9921),
(C) Irradiated to 2 Gy and stored at 25°C (R^2 = 0.9911),
(D) Irradiated to 2 Gy and stored at 5°C (R^2 = 0.9913).

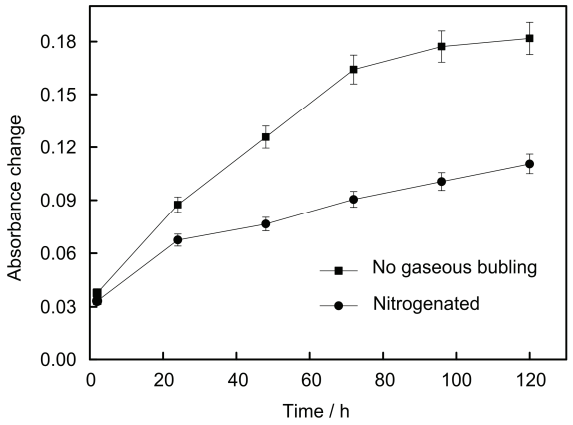

Fig. 4 Absorbance change of the non-irradiated FPGX gel dosimeters prepared with or without N_2 bubbling.

The impact of nitrogen on stability of the FPGX hydrogel dosimeter before irradiation was studied with the samples prepared in different atmospheric conditions. Results of the solution mixtures stirred with or without oxygen-removing N_2 bubbling are given in Fig.4. The sample with nitrogen bubbling was stabler, as the oxygen results in oxidation of Fe^{2+}. Therefore, purging the dosimeter by N_2 is necessary.

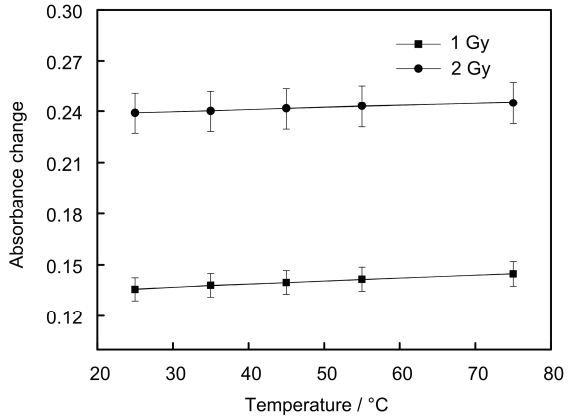

Fig 5. Absorbance change of the irradiated FPGX dosimeters measured at different temperatures.

Absorbance changes of the FPGX dosimeters irradiated at (25±2)°C and measured at 25°C, 35°C, 45°C, 55°C, and 75°C are shown in Fig. 5. The results did not seem to suggest any effect of the dose-reading temperature.

In conclusion, FPGX hydrogel dosimeters prepared by chemical crosslinking are stable under common application conditions in terms of storage time and temperature. The nitrogen purging is necessary to prepare FPGX hydrogel dosimeters of good stability.

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