Verification of a non-diffusing gel dosimeter

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Abstract. An evaluation of the diffusion coefficient of a genipin-gelatin gel dosimeter was carried out by fitting an inverse square root function to image profile data. A comparison was made with a Fricke-gelatin-xylenol orange (FGX) gel dosimeter, in which the ions are known to diffuse. The diffusion coefficient for this FGX gel, consisting of 0.5 mM ferrous ammonium sulphate, 50 mM sulphuric acid, 0.15 mM xylenol orange and 3 % by weight gelatin was 0.70 ± 0.05 mm² h⁻¹ at 5 Gy. The genipin-gelatin gel consisted of 50 μM genipin, 4 % by weight gelatin and 100 mM sulphuric acid. The fitted parameter that is proportional to the diffusion coefficient did not significantly change over time, demonstrating that this genipin-gelatin gel is a non-diffusing dosimeter.

1. Introduction

Polymer gel dosimeters are manufactured from radiation sensitive chemicals, which upon irradiation polymerize as a function of the absorbed radiation dose [1]. These gel dosimeters which record the radiation dose distribution in three-dimensions (3D) have specific advantages when compared to one-dimensional dosimeters and two-dimensional dosimeters [2]. These 3D dosimeters are radiologically soft-tissue equivalent [3] with properties that may be modified depending on the application. The 3D radiation dose distribution in polymer gel dosimeters may be imaged using magnetic resonance imaging (MRI) [4, 5], optical-computerized tomography (optical-CT) [6, 7], x-ray CT [8, 9], ultrasound [10-14] or vibrational spectroscopy [15, 16].

Gelatin-based gel dosimeters are made from radiation-sensitive materials that change as a function of absorbed dose, stabilised in a gelatin matrix. The resulting dose distribution within the gel volume needs to be spatially stable for a time sufficient to allow measurement of the dose distribution in three-dimensions (3D). A limiting feature of any gel dosimeter infused with radiation-sensitive species is the diffusion of those species in regions of dose gradients within the gel. This effect will eventually lead to a loss of 3D dosimetric information in the material.

In Fricke-infused gels, the radiation-sensitive ferrous ion (Fe²⁺) is oxidised upon irradiation to a ferric ion (Fe³⁺), and these ions will diffuse. Addition of the chelating agent, xylenol orange (XO), which binds to Fe³⁺ and forms a larger molecule, can reduce the diffusion coefficient [17-19]. It has
been reported that a genipin-gelatin gel does not diffuse post-irradiation [20]. Diffusion in a new genipin-gelatin gel dosimeter formulation [21] was quantitatively evaluated and compared with the diffusion coefficient determined in a Fricke-gelatin-xylenol orange (FGX) gel dosimeter, in which the ions are known to diffuse across dose gradients.

2. Methods
Genipin gel was prepared according to a new formulation [21] made from 50 μM genipin (078-03021, Wako Pure Chemical Industries), 4% by weight gelatin from porcine skin (300 bloom, G2500, Sigma-Aldrich) and 100 mM sulphuric acid (102761C, BDH) in the final gel. FGX gel was prepared according to a previous formulation [22] and the final gel consisted of 0.5 mM ferrous ammonium sulphate (09719, Fluka), 50 mM sulphuric acid (102761C, BDH), 0.15 mM xylenol orange (XO) (398187, Sigma-Aldrich) and 3% by weight gelatin from porcine skin (300 bloom, G2500, Sigma-Aldrich). All gels were pipetted into 1 cm UV-grade methacrylate cuvettes (Z188018, Sigma-Aldrich) and capped with disposable plastic caps (9020, Starna).

All gels were brought to room temperature prior to recording baseline transmission images using a modified flatbed optical scanner (Canon, MX850). Image processing settings were disabled in the scanner software to obtain as closely as possible, the raw image data in tagged image file format (tiff). The cuvette was placed in the centre of the scanner platen where the standard deviation in the noise of a background image was 0.3%. Profile slices for each sample were averaged from up to 10 consecutive images to improve the signal to noise ratio.

In order to create a sharp dose boundary, cuvettes were attached to the depleted uranium (DU) trimmer bars of a jaw collimator on an Eldorado-6 Atomic Energy of Canada (AECL) cobalt-60 therapy unit so that approximately half of the gel was shielded by the DU and the remainder exposed to the radiation beam at a dose rate of approximately 0.5 Gy min⁻¹. The gels were exposed to the source for a dose sufficient to clearly define a dose boundary. Three genipin-gelatin cuvettes were irradiated for 90 minutes and one FGX cuvette was irradiated for 10 minutes.

Image profiles were taken at various times after irradiation. The temperature during all measurements was 22 ± 1 °C. The average baseline image profile was subtracted from all post-irradiation profiles to obtain the raw data as relative pixel values. As we postulated that diffusion would not be observed in this genipin-gelatin gel dosimeter [20], a relatively simple technique was chosen to evaluate the diffusion coefficient. Data were fitted to an inverse square root function (equation 1). Terms used are analogous to the equation defined previously [19].

\[
dA = dA_0 + \frac{1}{2} (dA_1 - dA_0) \left( 1 + \frac{x}{\sqrt{x^2 + n}} \right)
\]

(1)

where \(dA\) is the measured value at the distance, \(x\), from the inflection point (the inflection point being at \(x = 0\)). \(dA_0\) is the minimum value of \(dA\) and \(dA_1\) the maximum value of \(dA\). \(n\) is a curvature parameter that varies inversely with the slope at the inflection point. \(dA_0\), \(dA_1\) and \(n\) are all fitted parameters. The slope of \(n\) versus time, multiplied by 0.212, is equal to the diffusion coefficient [23].

3. Results and discussion
Image profile data were fitted to the inverse square root function given in equation 1. For the FGX gel, a change in the slope at the inflection point over time can be seen in the fitted curves (figure 1), as a result of ion diffusion. Plots in figure 1 are also indicative of the lower dose sensitivity of the genipin-gelatin gel as compared to the FGX gel.

The curvature parameter, \(n\), increased linearly with time (figure 2a), and the diffusion coefficient was 0.70 ± 0.05 mm² h⁻¹ at 5 Gy. In the genipin-gelatin gel, \(n\) did not significantly change over the 45 h following irradiation (figure 2b). This confirms that there is no significant diffusion occurring in these genipin gels.
Figure 1. Fitted curves of image profiles of FGX gel (upper curves) and genipin-gelatin gel (lower curves) obtained at various times.

Figure 2a. The diffusion coefficient in this FGX gel was calculated from the slope of the linear fit of $n$ versus time and was $0.70 \pm 0.05 \text{ mm}^2 \text{ h}^{-1}$ at 5 Gy.

Figure 2b. The curvature parameter, $n$, did not significantly change over time for the genipin-gelatin gel dosimeter. Plotted are the average values (error bars indicate one standard deviation) of measurements of three samples from the same batch of gel.

4. Conclusion
The effect of diffusion on a genipin-gelatin gel dosimeter was evaluated and compared with the known diffusion in an FGX gel dosimeter. Genipin forms blue pigments as a result of cross-linking some of the amino acids that make up gelatin. The genipin-gelatin gel therefore differs from other gel dosimeters infused with radiation-sensitive species (such as Fricke-based gels) in that it will not diffuse in regions of dose gradients. A clear advantage of this genipin-gelatin gel over other types of hydrogel dosimeter systems is that this gel is non-diffusing. Other influences that have a more significant impact on this genipin-gelatin gel dosimeter in evaluating its usefulness for 3D dosimetry
include the dosimetric sensitivity, measurement uncertainty, stability and sensitivity to light and heat [24, 25].

5. References
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