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A diffusion-free and linear-energy-transfer-independent nanocomposite Fricke gel dosimeter

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HIGHLIGHTS

- We report a new magnetic-resonance-imaging based nanocomposite Fricke gel dosimeter.
- No diffusion of the radiation products was observed during nine days after the irradiation.
- Gel response faithfully reproduced the carbon beam depth-dose distribution.
- The NC-FG dosimeter exhibited a good linearity up to 800 Gy and suppression of LET effects.

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ABSTRACT

We report a new magnetic-resonance-imaging (MRI) based nanocomposite Fricke gel (NC-FG) dosimeter system, which is free from two main drawbacks of conventional Fricke gel dosimeters, namely, the diffusion of the radiation products and the linear-energy-transfer (LET) dependence of the radiation sensitivity when used for ion beams. The NC-FG dosimeter was prepared by incorporating 1% (w/w) clay nanoparticles into deaerated Fricke gel. We have dosimetrically characterized the NC-FG by using MRI measurements after irradiation with a monoenergetic 290 MeV/nucleon carbon beam. No diffusion of the radiation products was observed during nine days after the irradiation. Moreover, its response faithfully reproduced the depth-dose distribution measured by an ionization chamber, which indicates the absence of the LET dependence. Also, the NC-FG dosimeter exhibited a good linearity up to 800 Gy.

1. Introduction

The ferrous sulfate (Fricke) solution has been used as a reliable chemical radiation dosimeter for more than eighty years (Fricke and Hart, 1966; Fricke and Morse, 1927). Gore et al. (1984) proposed the addition of a gel matrix to the aqueous Fricke dosimeter in order to stabilize the spatial information of radiation-induced oxidation, which can be probed with magnetic resonance imaging (MRI). This has pioneered modern gel dosimetry (Baldock et al., 2010; Schreiner, 2004), a technique that records three dimensional (3D) dose distribution in tissue-equivalent gels.

One of the main drawbacks of Fricke gels, with respect to polymer gel dosimeters, is the diffusion of the ferrous and ferric ions despite the presence of the gel matrix, which eventually destroys the information on dose distribution (Penev and Mequanint, 2013). Another drawback is the decrease in radiation detection sensitivity with the increase in linear energy transfer (LET), which hinders absolute dose determination when used for ion beams. The LET dependence is not unique to Fricke gels but common for virtually all types of 3D dosimeters, as well as for film, scintillation, and semiconductor dosimeters (Karger et al., 2010).

In this paper, we report the successful removal of both of these limitations. We have recently shown that nanoclay addition can suppress radiation product diffusion in dichromate gel dosimeters

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(Maeyama et al., 2012; Maeyama et al., 2013; Taylor et al., 2013). Inspired by this, in the present study, we incorporated clay nanoparticles called Laponite XLG (Rockwood, 2013) into Fricke gel dosimeters. This nanocomposite Fricke gel (NC-FG) dosimeter succeeded in complete suppression of diffusion. Surprisingly, we also found that the NC-FG dosimeter exhibited the response almost independent of LET. Further, its radiation response was nearly linear up to at least 800 Gy.

2. Experimental

2.1. Sample preparation

Sample of the NC-FG and the Fricke xylenol orange gel (FXG) dosimeters were used in this study. The NC-FG was composed of 1% (w/w) nanoclay (synthetic hectorite, or Laponite XLG; Rockwood Ltd), 3% (w/w) gelatin (300 g Bloom from porcine skin; Sigma-Aldrich), 1 mM ammonium iron(II) sulfate and 50 mM perchloric acid. The procedure for the preparation of NC-FG was as follows: first, the ultra-pure water was exposed to N_2O gas via 30 min bubbling to exclude dissolved oxygen. Subsequently, under stirring, gelatin and Laponite XLG were added to this ultra-pure water, followed by heating until dissolution to obtain a uniform dispersion state. Finally, 5% (w/w) aqueous Fricke stock solution (Fricke and Hart, 1966) was added at around 40 °c. Thus the prepared NC-FG was sealed into color comparison tubes, made of Pyrex glass (Iwaki Glass Co), as shown in Fig. 1 and was refrigerated to gelation for a day after preparation.

The FXG was prepared as previously described in the literature (Kron et al., 1997; Rae et al., 1996) using 5% (w/w) gelatin, 1 mM ammonium iron (II) sulfate, 50 mM sulfuric acid and 0.5 mM xylenol orange.

2.2. Irradiation

The irradiation experiments were performed at Biological Irradiation Port of Heavy Ion Medical Accelerator in Chiba (HIMAC), National Institute of Radiological Sciences (NIRS). A carbon ion beam at 290 MeV/nucleon with an irradiation field having $\pm 5\%$ lateral dose uniformity within a diameter of 10 cm was used. The radiation doses on the surface of the samples were controlled by the dose monitor that is an ionization chamber and located in the upper beam line (Kanai et al., 2004; Torikoshi et al., 2007). The monitor unit (MU) value was calibrated by using the Markus ionization chamber at the same position of samples. The depth-dose distribution of a carbon-ion beam in this system is reported in the literature (Kanai et al., 1999). The dose rate on the incident surface of the present experiments was 7–8 Gy/min. Simultaneous irradiation of multiple gel samples was performed from the bottoms of the color comparison tubes in the radiation field. The irradiation dose is summarized in Table 1.

2.3. MRI measurements

An 1.5T MRI scanner (Intera Achieva 1.5T HP Nova Dual Gradient, Philips Medical Systems, Best, The Netherlands) was used for the measurement of these samples. The longitudinal MR relaxation rate ($R_1 = 1/T_1$) of the samples was evaluated by using a turbo mixed sequence (Baldock et al., 1998; Denkleef and Cuppen, 1987). The conditions of the T_1 measurements were: TR = 2260 ms; $TE_1 = 19$ ms $TE_2 = 100$ ms; TI = 500 ms; ETL = 12; pixel spacing = 0.78 mm. The elapsed days after the sample irradiation for the MRI measurements are summarized in Table 1.

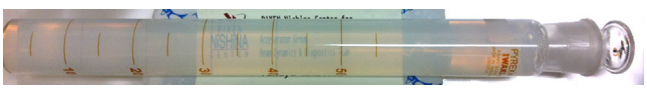

Fig. 1. Photograph of a deaerated NC-FG gel dosimeter.

Table 1
Irradiation surface doses and time of measurements for all gel dosimeters.

	Dose [Gy]	Elapsed days
NC-FG	200, 400, 600, 800	3, 9
FXG	0, 30	7

3. Results

3.1. Characteristics of NC-FG

An example of the R_1 ($1/T_1$) distributions measured for the FXG and the NC-FG irradiated with a 290 MeV/nucleon carbon beam is shown in Fig. 2. Fig. 2(a) and (b) refer to the FXG at 30 Gy and the NC-FG at 200 Gy, respectively. The NC-FG showed a very sharp peak near 140 mm, compared to the FXG. Given that the bottom of the color comparison tube which corresponds to the irradiation surface was not exactly flat, the distribution appears slightly curved in the y-axis direction. Further, the black part at the rightmost region (high region of R_1) represents the contamination of oxygen from the glass cap, causing autoxidation; however, it was confirmed that this black part did not spread thereafter.

Fig. 3 shows the R_1 increment ($R_1 - R_1(0)$) of the NC-FGs after irradiation with 200, 400, 600, 800 Gy. The R_1 value at the region with almost no dose contribution after the peak of each sample (around 190 mm), is used as $R_1(0)$. From Fig. 3, similar sharp distributions can be seen when the radiation dose was increased from 200 Gy to 800 Gy. The conventional Fricke gel dosimeters has a saturated dose response at 100 Gy (Schulz et al., 1990) and for the Fricke aqueous dosimeter this is reported to be 400 Gy (Matthews, 1982); above these values the linearity of dose response is lost due to the lack of Fe(II). Despite the very high dose at 4 kGy in the Bragg peak region, the peak observed for the 800 Gy incident dose shows the very sharp distribution similar to the 200 Gy incident dose, indicating that the progression of oxidation reaction of Fe(II) for absorbed dose is smaller than the conventional Fricke dosimeters.

In order to evaluate the dose dependence of the NC-FG, the $R_1 - R_1(0)$ values at various penetration depths were plotted in Fig. 4 as a function of the incident dose. The peak position near 141 mm for each sample differed to a small degree within MRI resolution (1 mm); hence, a minor correction was introduced in the direction of the penetration depth to adjust the peak position of each sample. From Fig. 4, a good linearity was confirmed at every position.

From the slopes of the dose-dependence curves in Fig. 4, the rate of R_1 increment (δR_1) per unit of entrance surface dose was evaluated and is plotted in Fig. 5. The δR_1 distributions after 3 days (dotted line) and 9 days (solid line) in Fig. 5 rendered very good consistency; it was found that the distribution did not change with time, implying that the diffusion of the product after irradiation was completely suppressed.

On the other hand, looking at the δR_1 distribution of the FXG gel dosimeter without the addition of nanoclay, there is almost no peak in its δR_1 distribution (right vertical axis in Fig. 5), presumably due to the reduction in dose response associated with increasing LET, and the diffusion of the radiation product.

The sensitivities of FXG and NC-FG near the entrance surface were 20 s^{-1} kGy^{-1} and 0.55 s^{-1} kGy^{-1}, respectively. It was found