Response of aqueous dichromate and nanoclay dichromate gel dosimeters to carbon ion irradiation

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Abstract. We have recently reported the significant reduction of radiation product diffusion by the incorporation of clay nanoparticles into dichromate gel (DCG) dosimeters. In this work, we investigate the influence of the nanoclay addition and gelation on the MRI $R_1 (1/T_1)$ image response of the dichromate dosimeter to the therapeutic carbon ion beam ($^{12}\text{C}^{6+}$ 290 MeV/u). The MRI $R_1$ distribution in the aqueous dichromate solution well reproduces physical dose-depth distribution with a high linear-energy-transfer (LET) efficiency. The nanoclay DCG dosimeters, on the other hand, exhibit composition-dependent LET efficiency degradation, while a sharp Bragg peak can still be detected. These results indicate that the nanocomposite gel addition may induce change in the radiation-induced reaction mechanism.

1. Introduction

Gel dosimetry [1, 2] is a suitable method for the validation of complex 3D dose distributions [3] encountered in contemporary radiotherapy including scanning-beam carbon ion radiotherapy [4]. However, implementation of polymer gels for carbon ion radiotherapy dosimetry has exhibited dose quenching around the Bragg peak [5-7]. Although the other gel dosimeter based on liquid chemical dosimeter such as Fricke and ceric sulfate gel dosimeter suffer from diffusion produced by irradiation [8, 9], it is expected that the dose response of the ceric sulfate gel shows relative insensitivity to LET changes [10, 11]. We have recently succeeded [12] in suppressing the diffusion in DCG dosimeters by adding clay nanoparticles, exploiting the property of the dichromate ion to adsorb to clay [13] and nanocomposite gel [14]. A liquid dichromate dosimeter based on radiation-induced reduction of $\text{Cr}_2\text{O}_7^{2-}$ is stoichiometrically liquid with the radiation-chemical yield of water radiolysis.
\[ G(Cr^{3+}) = 2 \times G\left(\text{Cr}_2\text{O}_7^{2-}\right) = \frac{1}{3}[G(\text{H}) + 2G(\text{H}_2\text{O}) - G(\text{OH})] \] (1)

The dose response of the dichromate gel dosimeter is expected to be unaffected by LET changes, since the OH and H yields cancel each other [15]. In this work, we investigate the influence of the nanoclay addition and gelation on the response of the dichromate dosimeter to the therapeutic carbon ion beam \(^{12}\text{C}^{6+}\) 290 MeV/u.

2. Material and methods

The aqueous dichromate solution prepared base on reference [16]. The nanoclay used here is a synthetic hectorite called Laponite XLG (Rockwood). Details of the chemical compositions of gel or liquid dosimeters are summarized in table 1. Beam of \(^{12}\text{C}^{6+}\) 290-MeV/u were provided from the Biological irradiation port in the Heavy Ion Medical Accelerator in Chiba (HIMAC) at National Institute of Radiological Science (NIRS). The dose-depth distribution were measured by ionization chamber using binary filter system consisting of plates made out of PMMA with different thicknesses corresponding to water thicknesses [17]. Liquid samples were irradiated in an array of 1-cm cuvettes at an entrance surface dose of 450 Gy with 56.33mmH\(_2\)O PMMA filter. The prepared gels were enclosed in Pyrex cylinder (Iwaki Glass Co., 9827TST: bottom thickness 2.1mm, diameter 32mm, length 230mm). Irradiations were undertaken from the bottom of Pyrex cylinder at an entrance surface dose of 470 Gy. The MR images were measured at 1.5-tesla (Intera Achieva Nova Dual, Philips Medical Systems, Best, The Netherlands) using Q-body coil. Calculation of the relaxation rates \(R_1 = 1/T_1\) was accomplished by using a “turbo-mixed” sequence (TR = 2260 ms, TE = 19 ms, TI = 500ms, ETL = 12, pixel spacing = 0.75 mm, NSA = 4).

| Table 1: Chemical composition of dichromate gel and liquid          |
|---------------------------------|----------------|-----------------|-----------------|----------------|
| Na\(_2\)Cr\(_2\)O\(_7\) mM    | HClO\(_4\) mM | AGAR w/v %      | LaponiteXLG w/v% |
| Gel1                           | 1              | 1               | 2.5             | 1              |
| Gel2                           | 1              | 25              | 2.5             | 1              |
| Gel3                           | 10             | 25              | 2.5             | 1              |
| Liquid                         | 2 mM K\(_2\)Cr\(_2\)O\(_7\) 50 mM HClO\(_4\) 0.1 mM AgClO\(_4\) |

3. Results and discussion

The MRI \(R_1\) distribution in the aqueous dichromate solution well reproduces physical dose-depth distribution with a high LET efficiency (figure 1). Since the product under irradiation in a 1-cm cuvette was homogeneous, we compared the total dose calculated from 1-cm length convolution of dose-depth distribution measured by ionization chamber and \(\delta R_1\) values. In the dose distribution, the dose ratio of the Bragg peak dose to the entrance surface dose is approximately 3, while the \(\delta R_1\) ratio of the value at the Bragg peak to the value at the entrance surface is approximately 2.5. The LET efficiency \(\varepsilon\) defined the following equation is approximately 0.8 at the Bragg Peak. From this result, we found that the aqueous dichromate solution dosimeter shows weak LET dependence.

\[
\varepsilon = \frac{[\delta R_1]_{\text{dose}} \text{ at the Bragg peak}}{[\delta R_1]_{\text{dose}} \text{ at the entrance surface}} \quad (2)
\]

The \(\delta R_1\) distributions of the nanoclay DCG are shown in figure 2 with dose-depth distribution. Since initial energy of ion was degraded by 2.1 mm Pyrex glass at entrance surface, the \(\delta R_1\) distributions were shown with adding 3.82mmH\(_2\)O depth. These results indicate that the dose response of nanoclay DCG is highly sensitive to compositional variation. Specifically, the sensitivity of the nanoclay DCG increases with increasing concentrations of perchloric acid or dichromate. We explicitly investigated concentration variations up to 500-mM dichromate. Gels containing high concentrations of dichromate
ion or perchloric acid take a long time to fix. As a result, inhomogeneous MRI images were obtained (not shown). In the dose-depth distribution, Dose at the Bragg peak indicates magnitude approximately 5 times higher than the surface dose. The $\delta R_1$ at the Bragg peak, in contrast, indicates magnitude approximately 1.5 times higher than the surface. We found that the nanoclay DCG response decreased with increasing LET. Especially, the LET efficiency was approximately 0.3 at the Bragg peak.

4. Conclusion
We have investigated the LET efficiency of the nanoclay DCG and aqueous dichromate solution using carbon ion irradiation (12C6+ 290 MeV/u). The aqueous dichromate dosimeter exhibits an LET efficiency (0.8) at the Bragg peak superior to that of the aqueous Fricke and coumarin solutions (< 0.5) [18, 19]. The results for the nanoclay DCG dosimeters indicate that the composition significantly influences the response and that the LET efficiency is degraded compared with that of the aqueous dichromate dosimeter.

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