Radio-fluorogenic nanoclay gel dosimeters with reduced linear energy transfer dependence for carbon-ion beam radiotherapy†

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The complex therapeutic dose distributions associated with new heavy-ion irradiation techniques require the development of 3D dosimeters; however, the precise measurement of dose distributions when using high linear energy transfer (LET) radiation remains a challenge. Specifically, the dosimeter sensitivity is typically decreased, and signal saturation occurs as a result of recombination reactions induced by high-density ionization and excitation. Thus, heavy-ion beam dosimetry necessitates a sensitivity correction for the LET being used. Consequently, we reported a magnetic resonance imaging-based 3D chemical gel ion-beam dosimeter for high LET irradiation that does not require the correction.[1)] However, several hundred Gy is required to perform measurements.

For improving the sensitivity, we examined the development of a new gel dosimeter based on fluorometry, which is the most sensitive detection method for radiation-induced products. Gel dosimeters comprise of a radiation-sensitive compound, a gelling agent, and water. In the case of low LET radiation, several types of radio-fluorogenic gel dosimeters have been reported to date, and we have prepared highly sensitive nanoclay radio-fluorogenic gel (NC-RFG)[2,3)] dosimeters from dihydrorhodamine 123 (DHR123) and nano-sized clay. The non-fluorescent DHR123 was oxidized to fluorescent rhodamine 123 (RD123) via exposure to ionizing radiation. The spatial information that describes the absorbed dose distribution was retained in the nanoclay gel matrix. Consequently, scanning spatial dosimetry can be performed using a fluorescent gel such as this because the fluorescence intensity increases linearly with increasing absorbed dose.

In the NC-RFG using DHR123, the dose-response or sensitivity of this gel dosimeters decreases with increasing LET, because RD123 is produced by oxidation reactions, primarily with OH radicals. Therefore, this study developed a method for reducing the effect of LET on NC-RFG dosimeters. Through the use of a new and optimal Fe^{3+} catalyst together with pyridine (Py) as a dispersant, more than 20 samples of NC-RFG with different preparation conditions were investigated. They were irradiated with 135 and 290 MeV/nucleon $^{12}C^{6+}$ ions accelerated by the RIKEN Ring Cyclotron and the Heavy Ion Medical Accelerator in Chiba, respectively. In this paper, only the preparation conditions after optimization are presented.

The prepared NC-RFG was deaerated with 1 mM Fe^{3+}, 100 mM Py, 100 μM DHR123, and 2 wt% Clay. The dose response of fluorescence intensity (I_{FL}) of irradiated samples was obtained via a fluorescence gel scanner. Figure 1(a) shows the IFL distributions obtained from the Fe^{3+}-Py-DHR123 NC-RFG using a 290 MeV/nucleon $^{12}C^{6+}$ ion beam for various entrance surface doses (ESDs). The distributions in (b) were determined by subtracting the pre-exposure distribution and dividing by the ESD. These plots demonstrate good agreement with one another, and the correlation coefficient (R^2) of I_{FL} versus dose was greater than 0.995 even near the Bragg peak, as shown in (c). Figure 1(d) presents a comparison of the relative δI_{FL}/ESD and absorbed dose distributions as evaluated through a standard method of dosimetry using an ionization camber (IC). These data confirm that the relative δI_{FL}/ESD distribution closely matched the dose distribution. Although the LET value increased to 266 eV/nm at the Bragg peak, the sensitivity was almost constant.[4)]

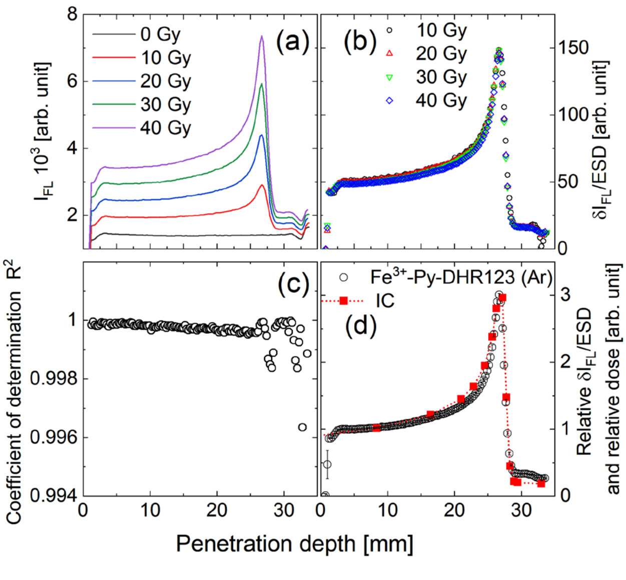

Fig. 1. Radiological properties of NC-RFG after irradiation with a carbon-ion beam ($^{12}C^{6+}$ 290 MeV/nucleon).[4)]

NC-RFG resulted in a significant reduction in the unwanted effect of the LET. The promotion of Fenton-like reactions decomposed H_2O_2 (the main product of the high-LET ion radiolysis of water) and thus en-

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hanced the oxidation reaction of DHR123. For the first time, this proposed dosimeter allows an evaluation of the depth dose profile when using a carbon-ion beam at a typical therapeutic dose level of several Gy without the need to perform sensitivity corrections based on LET.[4)]

References
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