Preliminary investigation of nanoclay-gel-based fluorescent gel dosimeters under carbon-ion beam

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High-precision radiotherapy that focuses high doses of radiation on tumors and decreases the amount of damage to healthy tissues is becoming popular. To validate the complex dose distribution, tissue-equivalent dosimetry with high spatial resolution in three dimensions is required. Gel dosimetry has attracted attention for dose verification in advanced radiotherapy. A gel dosimeter is prepared from a radiation-sensitive compound and a gelling agent. After a radiation-induced chemical reaction, the gel retains the same spatial distribution, and the 3D dose distribution can be evaluated by 3D gel scanning. Gel dosimeters are categorized according to the characterization method used the nuclear magnetic resonance (NMR) measurement method, $^{1-5)}$ absorptiometry $(X-ray^{6})$ or visible light^{7,8}), and fluorometry. The sensitivity of detection by fluorometry $^{9-11}$ is approximately 1,000 times greater than that by visible-light absorptiometry because fluorescence can be detected against a low background, isolated from excitation light. In contrast, absorptiometry measures transmitted light relative to high incident light levels at the same wavelength. The measurement methods using NMR and X-ray computed tomography (CT) also show limited detection sensitivity, and the dose of irradiation products must be at least several Gy for successful detection. In contrast, mGy-level dose can be detected by fluorometry. Thus, gel dosimeters employing fluorometry are expected to be the most highly sensitive type of gel dosimeter.

In a recent study, we reported a nanoclay-gel-based dosimeter that uses fluorescent dyes for X-ray irradiation. Although most gel dosimeters are prepared using organic gelling agents, such as gelatin, sensitivity can be enhanced by introducing inorganic nanosized clay as the gelling agent.⁹⁻¹¹) There are two types of such agents. The first is nanoclay-based radio-fluorogenic gel (NC-RFG), in which a fluorescent dye is produced when a non-fluorescent dye, such as coumarin-3-carboxylic acid (CCA), reacts with radicals formed from the radiolysis of water. The second type of gel is nanoclay-based radio degradation fluorescence gel (NC-RDG), in which a non-fluorescent dye is produced when a fluorescent dye, such as rhodamine 6G (R6G) or 7-diethylamino-4methylcoumarin (7D4MC), reacts with radicals. In this study, nanoclay-gel-based fluorescent gel dosimeters were applied to heavy-ion-beam irradiation for preliminary investigation.

Fluorescent gels were prepared using nanoclay as the gel, deionized water, and a fluorescence dye or probe. Two types of gel dosimeters prepared for this study. One was prepared from 2 μ M 7D4MC and 2.5-wt% nanoclay as follows. First, clay dispersion stock was prepared by



Fig. 1. Scanned image of fluorescent gel after irradiation with 135 MeV/nucleon ${}^{12}C^{6+}$ (entrance surface dose = 12 Gy).

mixing 23.35 g of ultra-pure water (Simplicity UV, Millipore Corp.) with 0.75 g of clay in a glass beaker at room temperature. A fluorescence probe stock solution (500 mL) was prepared by mixing 1.14 mg of 7D4MC with water. The clay dispersion stock (24.1 g) and fluorescence probe stock solution (5.9 mL) were then introduced to a 50 mL screw-capped glass vial and mixed using a rotation/revolution vacuum mixer (V-mini300, EME Corp., Japan) for 10 min at 1200/600 rpm and -90 hPa gauge pressure to obtain a uniform dispersion.

The other was prepared from 100 μ M dihydrorhodamine 123 (DHR123) and 2.5-wt% nanoclay using a rotation/revolution vacuum mixer with the same procedure. Finally, prepared gels were enclosed in a $100 \times 100 \times$ 10 mm plate. The prepared samples were irradiated using ¹²C⁶-ion beams (135 MeV/nucleon) accelerated by the RI Beam Factory (RIBF) at RIKEN. The irradiated samples were scanned using a fluorescent gel scanner.

The irradiation changes in fluorescence intensity with irradiation were observed, as shown in Fig. 1. The left image shows the entrance surface. In 7D4MC fluorescent gel, the fluorescence intensity decreases with increasing dose, and in DHR123 fluorescent gel, the fluorescence intensity increases with increasing dose. The detailed results are currently being analyzed, and some results will be submitted.

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