Two dimensional Raman mapping with respect to carbon bonds of radiochromic films: An approach to micro-dosimetry

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1. Introduction

Radiochromic films with an optical scanner are widely used as a useful dosimetry method in clinic area and provides good spatial resolution. Although, optical density based on light transmission provides proper measurement as a dosimetry, more precise methodology for enhanced dose contrast and high spatial resolution would be required for measurement of micro-dosimetry in cases such that low level energy is locally deposited in a sub-cellular size.

Raman spectroscopy usually provides fingerprints of chemical component species and molecular motion. Raman peak intensity can be quantified as dose changes. Using that Raman peak intensity is proportional to the electric field intensity of incidence beam and the concentration of compounds, the dose trend would have the linearity with the concentration change of radio-active compounds [1].

Raman spectroscopy has been applied to be utilized as a dosimetry in our group in the previous study[2]. Then, laser effect and film homogeneity issues were required to be overcome.

Two dimensional scan method was adapted to reduce measurement uncertainty since Raman cross-section is very sensitive to atomic bonds concentration and a large number of point measurements would guarantee reliable data group.

2. Methods and Results

In this section some of the techniques were used to detect Raman peak intensity as described.

2.1 Unlaminated Radiochromic Film

Special orders of Gafchromic unlaminated EBT3 films were required [3]. The intensity of Raman scattered light is about 0.01% of applied light and it is extremely hard to detect. Therefore, protect layers of radiochromic films are to be removed for more sensitive measurement. Active layer has the depth of about 30 micrometers and laser focusing should be performed in a very careful manner to measure exactly.

Especially, carbon atomic bonds are well known for generating strong Raman signal and EBT3 films feature that the concentration of carbon double and triple bonds change due to polymerization process.

2.2 Radiation delivery by linear accelerator

Photon irradiation of 6MV energy provided by therapeutic linear accelerator belongs to radiation oncology department in Seoul National University Hospital. Radiation delivery into the films was performed with the purpose of distinct Raman peak intensity difference by distinct dose difference.

Relative analysis was conducted through the way in that two dose contrast between non irradiated and irradiated part in one film sample was compared each other by split with the same laser focusing for better reliability.

For the clinic reference condition, 1 Monitor Unit (MU) is defined as a measure of machine output in radiation therapy. Irradiated output unit was measured by unit of MU. In this study, 1 MU is set up to generate 1 cGy for source to surface distance of 100 cm with a field size of 10 cm by 10 cm.

When preparing samples which has two split dose contrast, shielding material of Pb was applied onto films to shield scattered electrons by jaws so that penumbra is able to be at least.

2.3 Raman Spectrometer

A Raman spectrometer (DongWoo Optron Co., Ltd.) located at Seoul National University Graduate school of Convergence Science and Technology was used in this study. The spectra were analyzed using software (Andor Solis). Object lens had focus length of 200 cm and numerical aperture of 0.70.

When the polymerization brings about the change of carbon bonds concentration, Raman peak's affected by three Raman basic peaks altogether. Following the hypothesis that carbon bonds connected polymers with longer wavelength have red-shift as much as from 20 cm^{-1} to 40 cm^{-1}, the intensity of scanned monomer's peak would be affected by neighboring increasing peaks

altogether since not only monomer's decrease and polymer's increase of triple bonds but also double bonds increase resulting from polymerization affect one another in the vicinity of neighboring Raman peaks. However, at the level of this study's dose range, peak's increase of red-shifted was not obviously observed. Therefore, we focused on the first peak around 2070 cm^{-1} to see whether this peak has dose tendency. Those red-shifted peak showed up clearly by laser polymerization.

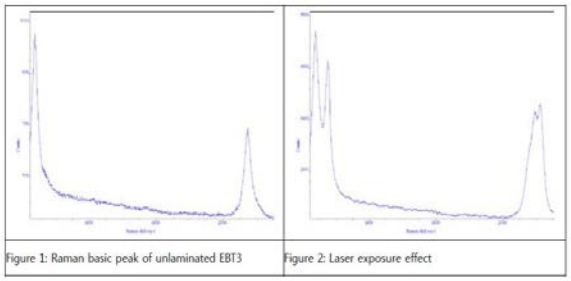

Fig. 1. Raman peak change due to laser polymerization

2.4 Raman 2D mapping

Mapping was performed by digitally controlled stages of the SG SP 26-100 model by Sigma Koki co.,ltd. Maple software was used to acquire two dimensional Raman peak distribution map, which is set up to reconstruct the distribution map of the maximum peak intensity within the scanned wave number range.

Specifically, the peak intensity of the wavenumber from 2065 to 2075 cm^{-1} was analyzed to reflect the polymerization concentration in light of radiation ionization.

Previous studies and more research revealed that laser affect Raman signal such that red-shifted Raman signal is generated by laser polymerization. In order to make these laser effect even for every exposure, even exposure of 1-2 seconds were controlled digitally.

Spatial resolution was enhanced up to 50 micrometers for most precise spatial resolution considering homogeneity issues of radio-active crystal component size of about 20 micrometers. However, the higher resolution, the longer mapping time's consumed.

Since Raman analysis is very sensitive to the focused Raman cross-section, relative analysis in that two split area, which has non-irradiated part and irradiated part, was compared each other for contrast.

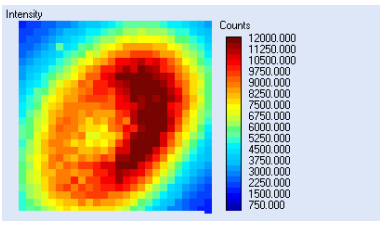

Fig. 1. 12.5 mm by 12.5 mm film reconstructed image by two dimensional mapping of Raman peak intensity for 10 Gy with the spatial resolution of 500 micrometers

Fig. 1 shows the result comparing with right irradiated split-side with non-irradiated left side in a contrast manner. Fig. 1 shows elliptical ring patterns in terms of not only polymerization degree but symmetrical physical curvature of films, which result in Raman cross-section. Irradiated right-split side shows higher counts (darker) than left comparatively especially for centered area.

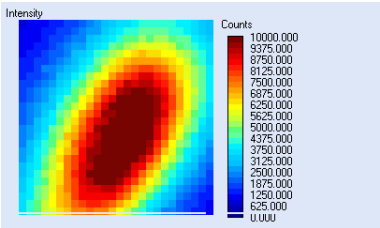

Fig. 2. 12.5 mm by 12.5 mm film reconstructed with no dose contrast but one totally irradiated film with output of 10 Gy with the resolution of 500 micrometers

Ring patterns confused the degree of polymerization quantity with another uncertainty factors. With a purpose of removing the ring patterns, films were fixed by applying tapes on the stage and scanned repeatedly to remove factors affecting the Raman cross-section such as geometrical curvature.

The contrast is only significant when compared with the non-irradiated film. Therefore, in order to get more distinct border of dose contrast was conducted by applying a line on the films with a color marker so that Raman signals for a marked line could be reconstructed as noise to be reflected as the border in the image.

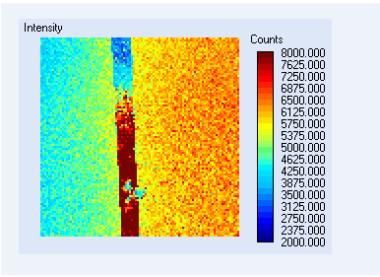

Fig. 3. 5 mm by 5 mm, central contrast by marked-split for 400 cGy with the resolution of 50 micrometers

The correlation between Raman peak's intensity and dose, which has not been shown evidently by the analysis of point-Raman signals, was distinct through the methodology of two dimensional analysis.

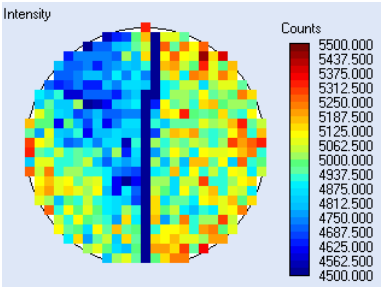

Fig. 4. 10 mm diameter mapping, central contrast for 10 cGy with the resolution of 100 micrometers. Although, there exists statistical uncertainty, low level radiation can be also d distinguished by acquiring a number of measurements.

3. Conclusions

The concentration in carbon double and triple bonds of radiochromic films would change by polymerization process. Thus, two dimensional analysis based on Raman mapping provides more reliable data in light of polymerization quantity due to radiation ionization than optical scanning. Its high spatial resolution (fifty micrometers) and low dose sensitivity (10 cGy) were demonstrated as a potential dosimeter. Raman analysis is expected as more precise analysis for micro-dosimetry in the future.

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