Transactions of the Korean Nuclear Society Autumn Meeting
Yeosu, Korea, October 25-26, 2018

A New Promising Approach for Radiochromic Film-Dosimetry at micrometer resolution using a Two Peak Ratio Method by Confocal Raman Spectroscopy

Rodrigo Hernández Millares [a], Geon Il Kim [b], Sung-Joon Ye [a,c*]

[a]*Program in Biomedical Radiation Sciences, Department of Transdisciplinary Studies, Graduate School of Convergence Science and Technology Seoul National University, Seoul 088826, South Korea*

[b]*Department of Nuclear Engineering, Seoul National University, Seoul 08826, South Korea*

[c]*Center for Convergence Research on Robotics, Advance Institutes of Convergence Technology, Seoul National University, Suwon 16229, South Korea*

Corresponding author: sye@snu.ac.kr

1. Introduction

Recent developments in radiation therapy techniques have heightened the need for micrometer resolution dosimetry. For instance, Stereotactic ablative radiotherapy, brachytherapy, radiobiology, and space industry require dose evaluation in regions with high gradients, at buildup and interface regions. Polymer gels and radiochromic films (RCF) have the spatial resolution, water equivalency, and accuracy. In this study, we used EBT-3 and EBT-XD radiochromic films. The active layer of Gafchromic™ EBT series films (Ashland Specialty Ingredients, Bridge Water, NJ) consists of diacetylene monomers in an ordered form, which on the application of UV light or high-energy radiation are polymerized and progressively darkens with increased radiation exposure [1]. There have been several investigations into dosimetric characterization of RCFs using an optical scanner and optical densitometer [2]. However, not many studies exist which covers the intrinsic spatial resolution of RCF. Christopher G Soares [3] mentioned the absolute limit of the RCFs is 0.75 μm which is the diameter of the chromophore. To achieve micrometer resolution using RCFs, it is essential to employ a high spatial resolution dosimetry system. Mirza *et al* [4] observed C≡C stretching band height of the diacetylene polymer and obtained a high spatial resolution of a few μm using a Raman spectroscopy. However, this method has a limitation because of the non-uniformity of RCFs. To minimize the dose variation of RCFs due to chromophore structure, axial stage movement, dust particles, micro-dent only measuring C≡C stretching band is insufficient [5]. By taking a two peak ratio method it is possible to reduce the dose variation. The main purpose of this study was to quantify the exposed radiation dose response in the micrometer level by measuring the ratio of C-C-C deformation band and C≡C stretching band height with reducing the uncertainty of RCFs. We obtained the calibration curves, dosimetric sensitivity, dose non-uniformity and effect of orientation of both EBT-3 and EBT-XD by using a two peak ratio method.

2. Methods and materials

2.1. Films and irradiation

Laminated EBT-3 (Lot #04051602) and EBT-XD (Lot #12101501) films were prepared and stored carefully to avoid possible exposure to the radiation as well as potential contamination of the active layer. EBT-3 film is composed of an active layer of 28 μm thickness, which consists of radiation sensitive diacetylene monomers. The active layer is compressed between two 125 μm thickness matte polyester substrates. EBT-XD has a same composition as the EBT-3, with the exception that EBT-XD has a 25 μm active layer and different chromophore's structure and size. The EBT-3 and EBT-XD films were cut into a 2×2 cm^2 size from the initial 20.32×25.4 cm^2 sheets. An arrow mark in the upper right corner was drawn for indicating the orientation of the film. The films were irradiated with a 6MV beam by a clinical linear accelerator in Daecheong hospital using the in-phantom method, being exposed to 0.3, 0.5, 1, 2, 4, 6, 8, 10, 15, 20, 30, 40, 50 Gy at a source to surface distance (SSD) of 100 cm.

2.2. Confocal Raman spectroscopy using immersion oil

Raman spectra of each film were measured by confocal Raman spectroscopy system (Dongwoo Optron Co.Ltd, Korea). A He-Ne laser(18mW) operating at 632.8nm was used as excitation source with an Olympus BX53 microscope and BX3-URA illuminator with U Plan SApo 100×objective of 1.40 numerical aperture for immersion oil (Mitutoyo Corp., Japan). The 150grooves/mm grating was selected to obtain the whole Raman shift range. To maximize a signal to noise ratio (SNR) and minimize laser exposure, parameters of entrance slit size, laser exposure time and accumulated spectra number was optimized for 100μm, 0.2sec, 10 respectively.

2.3. Raman mapping

We at Seoul National University have developed a Raman mapping technique to reduce the variation of C≡C stretching band intensity [4]. We selected a region of interest (ROI) of 100×100 μm^2 in each film randomly with a spatial resolution of 10 μm in the same way. By doing this, 121 Raman spectra with the Raman shift range from 50cm^{-1} to 4200cm^{-1} were prepared to analyze. C-C-C deformation bands and C≡C stretching bands were observed at 696 cm^{-1} and 2058 cm^{-1} respectively in each spectrum. Those two bands in each Raman spectrum were preprocessed with band selection, baseline removal and smoothing to obtain peak intensity. After preprocessing, the subtracted peak was fitted by a Gaussian function to calculate the band height. 121 band heights of C-C-C deformation band and C≡C stretching band were averaged and a two peak ratio value was obtained by dividing averaged C≡C stretching bands height by averaged C-C-C deformation bands height in all dose range as:

$$\text{Ratio}=\frac{BH_{C\equiv C}}{BH_{c-c-c}} \qquad (1)$$

2.4. Calibration curve, dose uniformity, dose sensitivity, dose range and effect of orientation

The Ratio (1) obtained for each dose level was used for drawing the calibration curves, afterward data points were fitted to an exponential function. The collected standard deviation (SD) and relative standard deviation (RSD) were used for quantification of the dose non-uniformity. Whereas for the case of dose sensitivity, the fitted curves previously obtained were normalized to compare both films. Independent two-sample T-test was done to verify the dose range for clinical usage. For data acquisition, the film was usually set on the mapping stage with landscape orientation, so that the arrow was on the upper right corner. But for the comparison of orientation, the arrow was set to be in the upper down right corner which means portrait orientation. For quantifying the orientation effect, EBT-3 and EBT-XD films up to 30 Gy were analyzed and the equation (2) was used with the corresponding uncertainties calculated through propagation error.

$$\text{OR}=\frac{Ratio_{6MV}(D,P)}{Ratio_{6MV}(D,L)} \qquad (2)$$

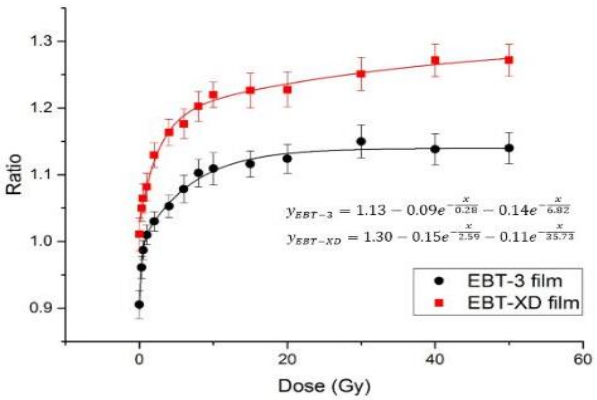
where $Ratio_{6MV}$ refers to the ratio obtained at the dose level D for the landscape and portrait orientation.

3. Results

3.1. Calibration curve, dose range, dose sensitivity and dose uniformity

Fig. 1(a) shows the dose response for Ratio (1). The saturation point found for the EBT-3 film after performing the corresponding statistical analysis at a significance level of 0.05 was 20 Gy while for EBT-XD it was 50 Gy. Figure 1(b) shows the normalization from the fitted curves shown in Figure 1(a) for sensitivity comparison purposes. It can be observed a higher sensitive response for the EBT-3 film.

a) Calibration curves



b) Normalized curves

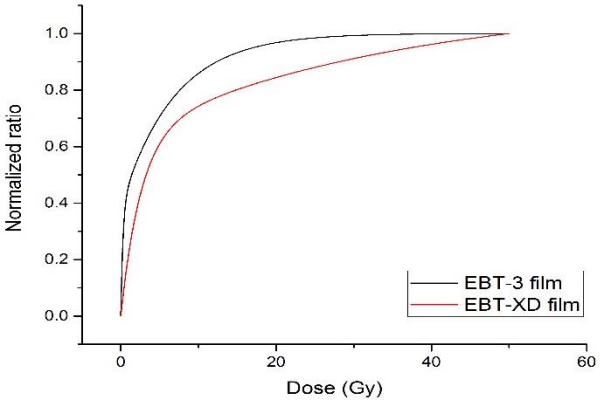


Fig. 1. (a) Calibration curves using the Ratio method up to 50 Gy. Error bars denote the standard deviation of the 121 Ratio values. (b) Normalization of fitted curves for the sensitivity comparison between the two films.

Table 1 and 2 indicate the RSD measured for data obtained with the Ratio method. Dose non-uniformity resulted to be less than 3% for all dose levels, indicating lower uncertainty of the dose distributed throughout the film. By analyzing the Raman spectrum in the C≡C stretching band, the typical values measured for the RSD are about 10%. Hence, using the Ratio method provides a significant improvement in dose non-uniformity of the film. A possible explanation for the different dose distribution across the film might be due to the stage micrometer uncertainty. Another cause could be due to an uneven structure in the micrometer level of the film. In addition, Table 1 and 2 show the RSD obtained by analyzing the C≡C band height, with the intention to compare between the dose uniformity response for the C≡C intensity and the Ratio method for EBT-3 and EBT-XD, respectively.

EBT-3				
Dose (Gy)	Mean	SD	RSD (%) Ratio	RSD (%) C≡C
0	0.906	0.021	2.3	12.7
0.3	0.961	0.016	1.7	13.5
0.5	0.987	0.014	1.5	11.8
1	1.010	0.015	1.5	10.5
2	1.030	0.015	1.4	11.7
4	1.053	0.017	1.6	11.0
6	1.079	0.021	1.9	12.1
8	1.103	0.021	1.9	14.2
10	1.110	0.024	2.2	13.4
15	1.116	0.020	1.8	10.0
20	1.124	0.022	1.9	11.7
30	1.150	0.025	2.1	11.0
40	1.138	0.023	2.1	10.4
50	1.140	0.023	2.0	11.3

Table 1. Measurements acquired from EBT-3 film. The mean and standard deviation (SD) were obtained after averaging the Ratio. An additional column with the C≡C analysis is shown to emphasize the improvement for the dose uniformity with the Ratio method.

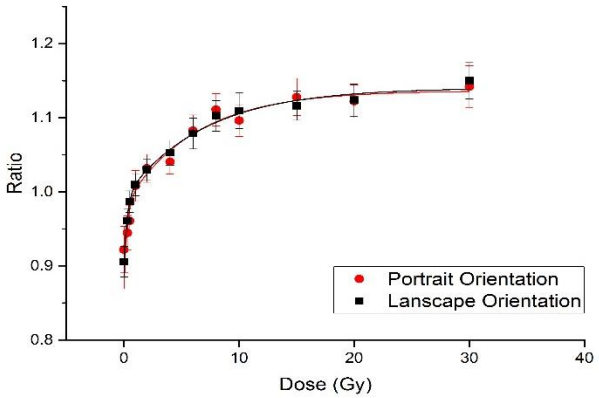
EBT-XD				
Dose (Gy)	Mean	SD	RSD (%) Ratio	RSD (%) C≡C
0	1.011	0.025	2.4	13.3
0.3	1.050	0.020	1.9	12.3
0.5	1.065	0.022	2.0	12.0
1	1.082	0.021	1.9	12.8
2	1.130	0.019	1.6	12.1
4	1.163	0.020	1.7	11.3
6	1.176	0.022	1.9	14.4
8	1.203	0.022	1.9	15.0
10	1.220	0.019	1.6	11.5
15	1.226	0.026	2.1	14.0
20	1.278	0.026	2.1	13.7
30	1.251	0.025	2.0	9.9
40	1.272	0.025	1.9	10.4
50	1.272	0.024	1.9	11.2

Table 2. Data collected from the EBT-XD film for the dose uniformity experiment using the Ratio method. As previously mentioned in Table 1, the RSD from C≡C analysis was added to highlight the improved performance of the Ratio method.

3.2. *Effect of Orientation*

In Fig.2 (a), the averaged values obtained through the Ratio method were fitted as an exponential curve for the landscape and the portrait orientation. The sensitivity of the EBT-3 films in different orientations was found to be less than 3%. Following the same tendency, EBT-XD showed a higher response in the landscape orientation and the variation due to orientation was less than 4%. Also, by observing Figure 2 (b), it is possible to notice the poor dosimetric performance of the EBT-XD in portrait orientation for doses below 1 Gy with the Ratio method. Therefore, it is not recommended to use the Ratio method for portrait orientation of EBT-XD film in an attempt to evaluate dose response.

a) Effect of orientation EBT-3



b) Effect of orientation EBT-XD

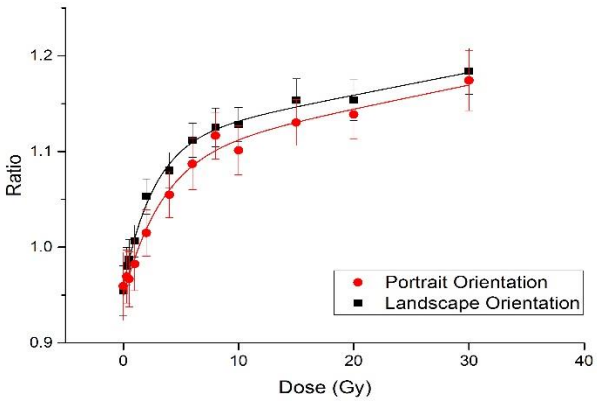


Fig.2 Dose response up to 30 Gy for different orientations (landscape and portrait) for (a) EBT-3 and (b) EBT-XD.

4. Conclusion

The results obtained through this work demonstrate the potential of applying the Ratio method for micro-dosimetry using Raman spectroscopy. The RCF structure's non-uniformity at a micrometer level is

compensated by obtaining the ratio between two different peaks of the Raman spectrum. EBT-3 is recommended to use up to 20 Gy, whereas EBT-XD is suitable for higher doses [50Gy]. EBT-3 shows higher sensitivity than EBT-XD. The ratio response for EBT3 and EBT-XD film in landscape orientation was more sensitive than those in portrait orientation for all doses. In the future, new experiments such as energy dependence, dose rate dependence and post-irradiation stability will be held on radiochromic films with the purpose of verifying the Ratio method as a potential method in micrometer dosimetry.

5. References

[1] S. Devic, N. Tomic, and D. Lewis, “Reference radiochromic film dosimetry: review of technical aspects,” Phys. Med. 32, 541-556 (2016).
[2] Hideharu Miura, Shuichi Ozawa, Fumika Hosono, Naoki Sumida, Toshiya Okazue, Kiyoshi Yamada, and Yasushi Nagata, “Gafchromic EBT-XD film: Dosimetry characterization in high-dose, volumetric-modulated arc therapy,” Applied Clinical Medical Physics, Vol 17, No.6, 312-322(2016).
[3] C. G. Soares, “Radiochromic film dosimetry,” J. Radiat. Meas. **41**, S100–S116 (2007).
[4] J. A. Mirza, H. Park, S Park, and S Ye, "Use of radiochromic film as a high-spatial resolution dosimeter by Raman spectroscopy," Med. Phys. **43**, 4520-4528 (2016).
[5] S. Bartzsch, J. Lott, K. Welsch, E. Bräuer-Krisch, and U. Oelfke, “Micrometer-resolved film dosimetry using a microscope in microbeam radiation therapy,” Med. Phys. **42**, 4069-4079 (2015).