Microstructure and dosimetric characterisation of delaminated film dosimeter under 12 MeV electron beam

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Abstract. Delaminated GAFCHROMIC™ EBT3 radiochromic films (RCFs) are specialised dosimeters that are gaining attention due to its many prospective advantages but are not commercially available. The purpose of this study is to fabricate the delaminated EBT3 RCFs, microscopically examine the structural response of the active layer (LiPCDA), and evaluate its dose-response in electron beam radiotherapy. The delaminated EBT3 RCFs were first made by carefully removing one of the two 125 μm polyester substrates using precision forceps. Next, the films were irradiated using a 12 MeV electron beam under full equilibrium conditions. Microstructure analysis via scanning electron microscope (SEM) was employed to look into the effects of radiation polymerisation on the LiPCDA crystals at 3000× magnification. To characterise the dose-response, the delaminated EBT3 RCFs’ optical density were analysed in Red-channel across a dose range of up to 5 Gy. The resulting calibration curve was then compared with the standard EBT3 RCFs’ curve for performance evaluation. Images from SEM revealed that the irradiated EBT3 RCF possesses a higher density of longer LiPCDA crystals (mode: ~8.00 μm) compared to the non-irradiated EBT3 RCF (mode: ~4.00 μm). The delaminated EBT3 RCFs, nonetheless, was found to produce a combined standard uncertainty of 6.62 %, which is inferior to its standard counterpart (3.62 %) and also the radiotherapy’s tolerance level (± 5 %). In sum, this study had successfully demonstrated the effects of radiation polymerisation on the active layer. However, the delamination process introduced a significant amount of uncertainty towards the dose-response. More efforts into delamination techniques and better dosimetry analysis are required to improve its usability in radiotherapy.

1. Introduction

Delaminated GAFCHROMIC™ EBT3 radiochromic films (RCF; Ashland Specialty Ingredients, Bridgewater, NJ, USA) are advantageous in strongly attenuated radiation modalities because the unprotected active layer can directly absorb radiation [1]. Among the application of this dosimeter is in low energy electrons detection, alpha radiation dosimetry, and also heavy particle beam calibration [1-4]. However, the purchasing of this custom dosimeter is costlier than the standard version. Therefore, many researchers have opted to fabricate their delaminated RCFs. One of the methods found in the literature was developed by Ng et al. [2]. The immersion of silicone-sealed standard RCFs into deionised water for 19 days before delamination was reported not to affect the dosimetric performance (evaluated through reflective optical density). Even so, the fabrication method used is not
time-efficient for on-the-spot measurements. Plus, the prolonged humidity exposure may cause surface texture damage on the RCFs’ polyester surface, which could affect the optical density sampling [5]. Another point that was not discussed accordingly was the lack of thorough uncertainty analysis on delaminated RCFs using flatbed scanner-based dosimetry. Thus, the purpose of this study is to fabricate delaminated EBT3 RCFs using a rapid and straightforward method, followed by microstructure and dosimetric performance characterisation under 12 MeV electron beam irradiation.

2. Materials and Methods

2.1. Radiochromic films
Standard EBT3 RCFs (Lot #: 05011702, Exp. date: May 2019) were chosen for experimentation. Its structural configuration consists of a 28 µm active layer compound (lithium salt of pentacosa-10, 12-diynoic acid, or LiPCDA diacetylene monomer crystals), sandwiched between two identical matte silica-coated polyester substrate of 125 µm. To create the delaminated films, the RCFs were first cut into small pieces of 1.0 × 1.2 cm² in dimensions. One of the polyester substrates was then carefully separated using a precision forceps, leaving the active layer exposed. The delaminated RCFs were handled according to the general RCF dosimetry procedures outlined by the AAPM TG-55 report [6].

2.2. Irradiation conditions
Irradiation of delaminated EBT3 RCFs using photon, proton, and 192Ir brachytherapy source has been reported in our previous paper [7]. As a continuation, 12 MeV electron beam was selected as the modality-of-choice, and irradiations were delivered using a PRIMUS™ linear accelerator unit (Siemens Medical Systems, CA, USA). All of the samples were positioned at a depth of maximum dose of 3.1 cm under tissue-equivalent gel bolus (CIVCO®, Osaka, Japan), using a 10 × 10 cm² beam field size at a source-to-surface distance of 100 cm. The total thickness of the backscatter solid water phantom (Nuclear Associates Inc., NY, USA) is 13 cm. Calibration was then conducted by exposing the delaminated RCFs to 10 dose levels from 0.5 Gy to 5 Gy. To minimise the statistical uncertainty in dose measurements, irradiation for each dose was repeated three times.

2.3. Microstructure characterisation
To investigate the EBT3 RCFs’ active layer structure response towards irradiation, scanning electron microscope (SEM) images were acquired from the delaminated films. Two delaminated RCF pieces were selected for the analysis, which is non-irradiated (0 Gy) and irradiated (4 Gy). The RCF pieces were firstly placed on its respective specimen holders with double-sided carbon tape and sputter-coated with a 6 nm gold element (Leica EM SCD005; Leica Microsystems, Wetzlar, Germany). Next, the samples were loaded into a field emission-SEM (Quanta™ FEG 450; FEI™, Netherlands) for secondary electron mode imaging, under the accelerating voltage condition of 5 kV to obtain the highest image resolution in radiation-sensitive polymers [8]. The final two images were then acquired in 3000× magnification and saved according to the manufacturer’s settings. Quantitative analysis of the structural response was carried out using open-source software (ImageJ v1.52i; NIH, Bethesda, MD, USA), whereby it was used to estimate the individual length of forty LiPCDA crystals per image.

2.4. Dosimetric characterisation
2.4.1. Digitisation. Film scanning was accomplished 72-hours post-irradiation. The EBT3 RCFs were scanned using EPSON® Expression® 10000XL flatbed scanner (Epson Seiko Corp., Nagano, Japan) in the following settings: transmission mode, colour positive film type, 24-bit RGB in Red-channel and digital image resolution of 150 dpi.

2.4.2. Film response and calibration procedure. Pixel values (PV) from the digitised RCFs were extracted using FilmCal software v2.4 (PTW-Freiburg, Freiburg, Germany). Three equally-sized region-of-interest (ROIs) were appropriately placed at the centre of the images to avoid any strain.
artefact at the edges (a total of nine ROIs across triplicate measurements). Changes in the optical density \((\text{netOD})\) and its reproducibility \((\sigma_{\text{netOD}})\) were defined by Devic et al. [9] as follows, in Equation (1) and (2):

\[
\text{netOD} = OD_{\text{irr}} - OD_{\text{non-irr}} = \log_{10} \left( \frac{PV_{\text{non-irr}}}{PV_{\text{irr}}} \right)
\]

\[
\sigma_{\text{netOD}} = \frac{1}{\ln(10)} \left( \left( \frac{\sigma_{PV_{\text{non-irr}}}}{PV_{\text{non-irr}}} \right)^2 + \left( \frac{\sigma_{PV_{\text{irr}}}}{PV_{\text{irr}}} \right)^2 \right)^{1/2}
\]

\(PV_{\text{non-irr}}\) is the averaged pixel value of non-irradiated RCFs and \(PV_{\text{irr}}\) is the averaged pixel value of irradiated RCFs. \(\sigma_{PV_{\text{non-irr}}}\) and \(\sigma_{PV_{\text{irr}}}\), on the other hand, are the standard deviations of \(PV_{\text{non-irr}}\) and \(PV_{\text{irr}}\) obtained from the mean in the ROIs, which also corresponds to film homogeneity uncertainty.

To establish the dose-response or calibration curve, OriginPro 2018 software (OriginLab, Massachusetts, USA) was used to fit the \(\text{netOD}\) datasets with Equation (3) [9]. \(B, C, \) and \(n\) were identified as the fitting parameters, and \(D\) is the measured absorbed dose in Gy:

\[
D = B \cdot \text{netOD} + C \cdot \text{netOD}^n
\]

Dose determination uncertainty or total scan uncertainty \((\sigma_{D_{\text{total}}})\), which consisted of the experimental uncertainty \((\sigma_{\text{exp}}; \) irradiation and scanning) and fitting uncertainty \((\sigma_{\text{fit}})\) was expressed mathematically in Equation (4) [9]:

\[
\sigma_{D_{\text{total}}} = \left( \left( \frac{\sigma^{2}_{\text{exp}}}{} \right) + \left( \frac{\sigma^{2}_{\text{fit}}}{} \right) \right)^{1/2} \left( \left( \frac{\sigma^{2}_{\text{netOD}}}{} \right) \right)^{1/2}
\]

\[
D_{\text{fit}} = \sigma_{\text{netOD}} \cdot \sigma_{\text{netOD}} = \left( \left( \frac{\sigma^{2}_{\text{netOD}}}{\sigma_{\text{netOD}}} \right) \right)^{1/2}
\]

\(\sigma_{\text{netOD}}\) and \(\sigma_{\text{netOD}}\) were identified as the uncertainty for fitting parameter \(B\) and \(C\), and \(D_{\text{fit}}\) refers to the absorbed dose, \(D\) calculated using the fitting function of Equation (3).

3. Results and Discussions

Figure 1 (a) and (b) shows the active layer image of non-irradiated and irradiated EBT3 RCF from the SEM imaging. The results for its respective microstructure characterisation were presented in the form of probability distribution histograms in Figure 1 (c) and (d). The red dotted line shown on the histogram illustrates the mode value for non-irradiated LiPCDA. The non-irradiated film possessed a mean LiPCDA crystal length of 5.63 µm ± 2.33 µm (mode: ~4.00 µm; 14/40 measurements). Upon 4 Gy irradiation, the crystals elongated to a mean length of 7.05 µm ± 2.91 µm, which was indicated by a shift in the probability distribution (mode: ~8.00 µm; 12/40 measurements). This result confirms the effect of photo-polymerisation, where irradiation causes the individual diacetylene monomer to grow in length via progressive 1, 4-polymerisation into acetylene polymer [1].
Figure 1. Active layer micrographs of (a) non-irradiated and (b) irradiated GAFCHROMIC™ EBT3 RCF in 3000x magnification setup. Quantitative analyses on LiPCDA’s microstructure response show that the probability distribution of irradiated EBT3 RCF (d) shifts to the right upon irradiation, indicating longer polymer crystals as compared to the non-irradiated EBT3 RCF (c).

Besides clinical radiation, other surrounding factors may also contribute to the polymerisation of LiPCDA, i.e., heat, UV radiation, and magnetic field [10, 11]. Therefore, it is important to always maintain a standard handling and calibration procedure to ensure that dose uncertainties due to unwanted polymerisation were kept at a minimum. This strict dosimetry practice applies in particular for delaminated EBT3 RCFs, as the exposed active layer might be more prone to the polymerisation triggers as opposed to standard EBT3 RCFs. On a separate note, it would be interesting to observe the microstructural response for different beam qualities and energies. Since EBT3 RCFs have been improved in terms of its energy dependence at the kilovoltage spectrum compared to its predecessors [12], we predict that the degree of elongation to be only slightly different across multiple energies.

Calibration curves for standard and delaminated EBT3 RCFs under 12 MeV electron beam irradiation were superimposed on the same plot in Figure 2. The $n$ parameter in Equation (3) was fixed to 2.0, while the fitting process was optimised using the non-linear Levenberg-Marquardt algorithm. Results of coefficient-of-determination (COD) and its adjusted form suggested that Equation (3) appropriately fits the netOD datasets from both RCF types (0.999 for standard EBT3 RCFs versus 0.997 for delaminated EBT3 RCFs). Anyhow, the calibration curve for delaminated EBT3 RCFs was not in union with the curve for standard EBT3 RCFs. This observation could be interpreted in a way that the delamination had compromised the dosimeter’s overall performance.
Figure 2. Calibration curve comparison for standard and delaminated GAFCHROMIC™ EBT3 RCFs in Red-channel. The error bars correspond to the netOD reproducibility uncertainties.

The dose uncertainty analyses for both RCF types are shown in Table 1. There are two uncertainty categories: Type A is determined by replicate measurements, while non-statistical procedures estimate Type B. A divisor of $\left( \frac{3}{2} \right)^{1/2}$ was considered in the film homogeneity and reading reproducibility parameter to take into account the triplicate measurement in Type A uncertainty [13]. Results from the uncertainty budget showed that the total scan uncertainty contributed to the highest uncertainty percentage in both standard and delaminated EBT3 RCFs, followed by film homogeneity and reading reproducibility. However, the dosimetric performance of delaminated EBT3 RCFs was found to decline significantly as its combined standard uncertainty is higher (6.62 %) than both standard EBT3 RCFs (3.62 %) and the radiotherapy’s dose accuracy requirement (5.00 %) [6].

This finding is most likely to be linked with the heightened noise signal found in the delaminated EBT3 RCFs’ scanned images, which originates from the defects incurred on the active layer surface [14]. There are many examples of delamination-induced defects, including, but not limited to, active layer stripping and scratches. To mitigate this issue, several post-irradiation techniques can be considered in the dosimetry, i.e., triple-channel analysis [14, 15] and image denoising filter [16]. The methods mentioned, however, was not included in the current study as the aim was to only characterise the basic dosimetric performance of delaminated EBT3 RCFs.

Table 1. Dose uncertainty budget for standard and delaminated GAFCHROMIC™ EBT3 RCFs.

| Parameter                        | Type of uncertainty | Standard Value (%) | Divisor | Relative value (%) | Value (%) | Divisor | Relative value (%) |
|----------------------------------|---------------------|--------------------|---------|-------------------|-----------|---------|-------------------|
| Total scan (Experimental and fitting) | B                   | 3.60               | -       | 3.60              | 6.58      | -       | 6.58              |
| Film homogeneity                 | A                   | 0.60               | $\left( \frac{3}{2} \right)^{1/2}$ | 0.35 | 1.15               | $\left( \frac{3}{2} \right)^{1/2}$ | 0.66 |
| Reading reproducibility          | A                   | 0.25               | $\left( \frac{3}{2} \right)^{1/2}$ | 0.14 | 0.49               | $\left( \frac{3}{2} \right)^{1/2}$ | 0.28 |
| Combined standard uncertainty    | ( $k = 1$; %)       | 3.62               |         |                   | 6.62      |         |                   |
4. Conclusion
The rod-like microstructures of LiPCDA found inside GAFCHROMIC™ EBT3 RCFs were observed via SEM imaging, post-film delamination. Irradiation of 12 MeV electron beam had caused the microstructures to form longer polymer chains through multiple radiation-induced chemical processes. As for the dosimetric performance evaluation, the calibration curve non-unity and high combined standard uncertainty presented in the delaminated EBT3 RCFs indicated a decline in performance compared to the standard EBT3 RCFs. Future studies utilising manually delaminated RCFs should take into account the many uncertainties caused by the defects on the films’ surface, before reaching into any conclusion regarding the quantified netOD or dose. On the other hand, more investigation is warranted in the future to develop better delamination protocols. Such effort would potentially strike a balance between the research cost and quality of dosimeter measurement.

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