Diffusion properties of a radiochromic hydrogel dosimeter

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Abstract. The aim of this study was to investigate the diffusion properties of a radiochromic hydrogel dosimeter based on leuco malachite green dye in a gelatine matrix. One half of each dosimeter was irradiated while the other half was left un-irradiated creating dose gradients over which diffusion could be investigated. Read-out of the optical response was performed with a high-resolution optical scanner. The dosimeters were found to exhibit a low diffusion rate but a high auto-oxidation level leading to a fading of the contrast in the dose response with time.

1. Introduction
The high conformity of modern radiotherapy techniques has motivated research into dosimeters that offer full three-dimensional dose measurements. Dosimeters capable of such measurements include both polymer and radiochromic dosimeters \cite{1, 2}. Radiochromic hydrogel dosimeters are promising tools within three-dimensional dosimetry since i) they are easy to make, ii) they are insensitive to oxygen, and iii) they possess good properties for optical read-out due to a non-scattering dose response. However, the accuracy of such dosimeters can potentially be deteriorated due to diffusion of the optical response. Diffusion in radiochromic hydrogel dosimeters has previously been reported in the literature for various hydrogel dosimeters \cite{3-7}, however to our knowledge only for a single dose and temperature. In order to obtain a broader description of the diffusion of the optical response, we have in this study investigated the diffusion properties of a radiochromic hydrogel dosimeter based on leuco malachite green dye in a gelatine matrix for a range of doses as well as a range of post-irradiation storage temperatures.

2. Materials and Methods

2.1. Dosimeter fabrication
The dosimeter used in this study was a radiochromic hydrogel dosimeter as proposed in Ref. 3. The chemical composition consists of the active component leuco malachite green dye (0.37 mM) which was dissolved in 80 mM trichloromethane while 5 mM trichloroacetic acid was added as an initiator. This solution together with the surfactant sodium dodecyl sulphate (50 mM) was mixed with a 6 % (w/w) aqueous gelatine solution to form a three-dimensional dosimeter. The surfactant made it possible to dissolve the non-polar leuco malachite green dye and the trichloromethane in the aqueous gelatine solution due to formation of so-called micelles. The gel was prepared in standard PMMA cuvettes (1x1x4.5 cm) and placed in a refrigerator after fabrication.
2.2. Irradiation and temperature
Irradiation of the dosimeters was performed with a dose rate of 14.6 Gy/min using x-rays from a linear accelerator set at 6 MV. The dosimeters were placed on the gantry center axis with one jaw covering half the dosimeter, creating a dose gradient in the dosimeter. The upper half of the dosimeters were then irradiated to doses of 20 Gy, 40 Gy, 60 Gy or 80 Gy while the bottom half was left un-irradiated. Irradiation was performed at room temperature while the dosimeters were stored in a refrigerator before irradiation and at a series of temperatures ranging from 5.4 °C to 29.8 °C after irradiation. The storage temperatures after irradiation were stabilized with a precision of ±0.5 °C.

2.3. Read-out of the optical response
In order to perform high resolution measurements of the optical response in three dimensional dosimeters, a high resolution optical scanner was built. The scanner was designed to measure the optical density of a volume dosimeter along the axis of a standard cuvette or similar dosimeter container. A schematic illustration of the scanner is shown in figure 1. The laser beam from a helium-neon laser (632.8 nm) is shaped vertically with the use of two cylindrical lenses (-5.8 mm and 150 mm focal length). Together with an iris this creates a vertical beam diameter of 8 mm. Horizontally, a cylindrical lens with 150 mm focal length focuses the beam to 0.102±0.005 mm beam waist, i.e. a FWHM of 0.120±0.006 mm, positioned in the middle of the dosimeter. This was a compromise between a narrow beam waist and limited beam divergence (i.e. a sufficiently large Rayleigh length). When a cuvette is scanned along its axis, the vertically extended, horizontally narrow beam will result in a high resolution along the measurement direction while integrating the signal perpendicular to the cuvette axis. The latter reduces uncertainties due to impurities in the dosimeter as well as on the cuvette. In addition, a spherical lens (f = 58 mm focal length) is positioned behind the cuvette at a 2/f distance from both the dosimeter and the detector in order to image the middle of the dosimeter onto the detector to maintain a high spatial resolution of the scanning system.

The dosimeter was placed on a motion stage motorized with a stepper motor with a minimum step length well below the resolution limit of the optical system. The laser intensity was measured with two detectors, with a detector placed after the dosimeter measuring the transmitted light, and a reference detector, in order to compensate for possible instabilities in laser power. Both the data acquisition (Data Translation DT9834) and the motorized stage were computer controlled, and in this study the dosimeters were translated 0.2 mm between each step. At each position, a data sequence was recorded and averaged.
Figure 2: Dose gradients from 40 Gy (left side of the gradient) to 0 Gy in dosimeters stored after irradiation at 5.4 °C (left graph) and 20.8 °C (right graph). The lowest curves in the two graphs were measured 2 hours and 1 hour after irradiation, respectively. The following curves from bottom to top were measured 3, 5, 7, 19, 25, 45, 68, and 143 hours after irradiation. The solid lines are fits to Eq. 1.

The dosimeters were measured with the optical scanner 1-2 hours before irradiation as well as several times after irradiation to quantify the change in dose gradient with time due to diffusion of the optical response.

2.4. Data analysis

The change in optical density due to the irradiation, i.e. the optical response, was found by subtracting the optical densities of the pre-irradiation measurement from the post-irradiation measurements. The diffusion properties of the dosimeters were then analysed by fitting the optical response, or change in attenuation coefficient, to

$$\Delta\alpha(x) = AO + s \int \Delta\alpha_{\text{irr}}(x') \cdot \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{(x-x')^2}{2\sigma^2}\right) dx,$$

where $AO$ is a second order polynomial expression describing auto oxidation of the leuco dye and $s$ is a scaling factor. The integral describes a convolution of the first post-irradiation measurement, $\Delta\alpha_{\text{irr}}$, and a normal distribution describing the change from $\Delta\alpha_{\text{irr}}$, i.e. changes in optical response due to diffusion.

3. Results

Figure 2 shows the optical response as a function of position in two dosimeters for a range of post-irradiation measurements including fits to Eq. 1. The left graph shows a dosimeter stored at 5.4 °C after irradiation while the right graph shows a dosimeter stored at 20.8 °C. Both dosimeters have received 40 Gy with a gradient down to 0 Gy. The lowest curve in each graph was used as $\Delta\alpha_{\text{irr}}$ when fitting all other curves for each dosimeter. These curves were measured 2.1 hours and 0.9 hours after irradiation, respectively. The following curves shows post-irradiation measurements with the top curve approximately 143 hours after irradiation.

As clearly visible on the graphs, the observed diffusion is low. Values for the broadening of the optical response, $\sigma$, as determined from the fits was found to be constant in time for both temperatures with mean values of 0.08±0.01 mm and 0.09±0.01 mm at 5.4 °C and 20.8 °C, respectively. In addition, no considerable change in $\sigma$ was observed in the remaining temperature range of 11.3 °C, 14.9 °C, 25.0 °C and 29.8 °C or in the remaining dose range of 20 Gy to 80 Gy. The overall mean across all measurements was found to be 0.073±0.003 mm.
A pronounced auto-oxidation of the optical response was observed within the time range of the measurements (figure 2). The auto-oxidation not only resulted in an offset in optical response but also a curvature through the dosimeter, described by a second-order polynomial expression (AO in Eq. 1).

4. Discussion
In this study we have investigated the diffusion properties of the dose response in a radiochromic hydrogel dosimeter using a high resolution optical scanner. Since the dosimeter used in this study contains the non-polar leuco malachite green dye in a hydrogel, the dye is initially expected to be captured within micelles formed by the surfactant. In the expected reaction upon irradiation, the leuco dye is transformed to a coloured dye that is charged. The coloured dye might therefore acquire sufficient polar characteristics that it may diffuse out of the micelles. This, however, seems not to be a pronounced effect since a very low diffusion of the optical response was observed in this study. The change in dose gradients with time due to diffusion was observed to be lower than the resolution of the scanner. The constant value for the broadening of the initial gradient, σ, which was obtained for all fits to Eq. 1 independent of time after irradiation, post-irradiation temperature or dose, is therefore attributed to a measure for the reproducibility of the optical scanner.

Diffusion of the optical response has previously been reported for radiochromic hydrogel dosimeters, however with the use of another surfactant than sodium dodecyl sulfate [3, 4]. In addition diffusion in dosimeters with the same chemical composition as in this study has been reported [5]. In that study a diffusion rate was observed when taking into account offsets due to auto oxidation, and the remaining change with time was then attributed to diffusion. This is contrary to our study, where the only visible effect within the time range of the experiments of 143 hours was due to auto-oxidation as well as a relative fading of the signal caused by irradiation. In addition, the auto-oxidation in our study varied through the dosimeters and had to be described by a second-order polynomial expression. Similar auto-oxidation behaviour was also observed in a set of un-irradiated dosimeters, however, with different offsets between the optical response measured in the irradiated dosimeters and the un-irradiated dosimeters.

5. Conclusion
In terms of diffusion of the optical response, the radiochromic hydrogel dosimeter investigated here was observed to be stable with a diffusion lower than the resolution of the optical scanner. However, the dosimeter showed a high auto-oxidation in addition to a relative fading of the optical response with time.

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7. References
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