Dose rate dependency of micelle leucodye 3D gel dosimeters

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Abstract. Recently a novel 3D radiochromic gel dosimeter was introduced which uses micelles to dissolve a leucodye in a gelatin matrix. Experimental results show that this 3D micelle gel dosimeter was found to be dose rate dependent. A maximum difference in optical dose sensitivity of 70% was found for dose rates between 50 cGy min\(^{-1}\) and 400 cGy min\(^{-1}\). A novel composition of 3D radiochromic dosimeter is proposed composed of gelatin, sodium dodecyl sulphate, chloroform, trichloroacetic acid and leucomalachite green. The novel gel dosimeter formulation exhibits comparable radio-physical properties in respect to the composition previously proposed. Nevertheless, the novel formulation was found to be still dose rate dependent. A maximum difference of 33% was found for dose rates between 50 cGy min\(^{-1}\) and 400 cGy min\(^{-1}\). On the basis of these experimental results it is concluded that the leucodye micelle gel dosimeter is still unsatisfactory for clinical radiation therapy dose verifications. Some insights in the physico-chemical mechanisms were obtained and are discussed.

1. Introduction
3D radiochromic dosimeters change color upon radiation. The radiochromic dosimeter investigated in this study changes color upon irradiation due to the oxidation of leucodyes by halogen radicals. In commercially available products such as Presage\textsuperscript{TM}, the leucodyes and halogens are dissolved in a polyurethane matrix [1]. In recently published papers by Jordan \textit{et al.} and Babic \textit{et al.} another approach was taken: In their proposed dosimeter the color dye and halogen are dissolved in a gelatin gel [2,3]. As the color dye and halogen do not readily dissolve in the gelatin hydrogel, the dye and halogen are embedded in micelles. The authors concluded that the micelle gel dosimeters are potential 3D dosimeters that had no dose rate dependence. The authors conclusion on dose rate dependence was based on a comparison of the shape of a gel measured dose depth curve for electron beams and a dose depth curve acquired with an ionization chamber. In our study in which the dose rate dependency of the leucodye micelle gels were investigated independently with photon beams, however a significant dose rate dependency was found. The dose rate dependency of different kinds of leucodye gel dosimeters were investigated, including the gel dosimeter proposed by Jordan \textit{et al.} [2]. A novel leucodye gel dosimeter is proposed that has a similar dose sensitivity and improved dose rate dependency in respect to the composition proposed by Jordan \textit{et al.} [2].

2. Materials and methods
2.1. Gel fabrication
The leucodye micelle gel dosimeter proposed by Jordan \textit{et al.} (further referred to as LMD1) was fabricated according to the fabrication procedure described in their publication [2] and poured into
small cuvettes (1x1x4.5 cm³, PMMA, Sigma Aldrich) and placed in a refrigerator overnight at 4°C. These cuvettes are further referred to as spectroscopic samples.

A novel composition of leucodye micelle gel is proposed that consists of 6% gelatin, 80 mM chloroform (CHCl₃), 50 mM sodium dodecyl sulphate (SDS), 5 mM trichloroacetic acid (CCl₃COOH) an 0.37 mM leucomalachite green (LMG) all dissolved in deionised water (further referred to as LMD2). Gelatin is dissolved in 60% (w/w) of total water volume at room temperature and is left to swell for 10 minutes. Thereafter the gelatin-water solution is heated to 50°C. The remaining 32% of total water volume is used to dissolve SDS and CCl₃COOH. CHCl₃ is used to dissolve LMG after which this solution is carefully added to the water-SDS-CCl₃COOH-solution. The solution is wrapped in aluminum foil to minimize any photochemical induced reaction. After cooling of the gelatin-water solution to approximately 40°C, the two solutions are added together and stirred for 15 minutes in a dark room. Finally the gel is poured into small cuvettes, small PMMA phantoms (dimensions 1x1x8 cm³) and a large PMMA phantom (dimensions 1x1x40 cm³).

2.2. Dose sensitivity
Spectroscopic samples were irradiated with doses ranging between 0 Gy and 60 Gy in steps of 5 Gy and between 60 Gy and 100 Gy in steps of 10 Gy. Afterwards all spectroscopic samples were placed in a refrigerator overnight at 4°C. The spectroscopic samples were read out approximately 12 hours post irradiation by use of an USB 4000 spectrometer (Ocean Optics, Dunedin, USA). The ∆OD dose plot was fitted in Matlab and the slope was used in further comparisons.

2.3. Dose rate dependence
The dose rate dependency for the LMD1 type and LMD2 type dosimeter is investigated for photon beams of 6 MV by changing the photon beam pulsation rate. Spectroscopic samples were irradiated at dose rates of 50 cGy min⁻¹, 100 cGy min⁻¹, 200 cGy min⁻¹ and 400 cGy min⁻¹ at isocentre.

2.4. Depth dose profiles
Depth dose profiles were measured for electron beams of 6 MeV at an SSD of 95 cm and field size 10x10 cm² (using an electron applicator and appropriate cut-out) for dose rates set at 50 cGy min⁻¹, 200 cGy min⁻¹, and 400 cGy min⁻¹ at isocentre. The position of the central axis of the small PMMA phantoms was along the beam direction. Measured depth dose profiles were rescaled to dose maximum for the PMMA phantom irradiated at 400 cGy min⁻¹. Ionization chamber measurements for a 10x10 cm² electron field of 6 MeV were obtained from the treatment planning system. Depth dose profiles were compared with depth dose profiles acquired with the gel.

A depth dose profile was measured for a photon beam of 6 MV at an SSD of 95 cm and field size 10x10 cm² for a dose rate set at 400 cGy min⁻¹ at isocentre. The position of the central axis of the large PMMA phantom was along the beam direction. The measured depth dose profile was rescaled to dose maximum. Ionization chamber measurement was performed with a 0.016 cm³ pinpoint ionization chamber (PTW 31016) and automated water phantom (PTW). The depth dose profile was compared with the depth dose profile acquired with the gel.

All PMMA phantoms were read out by the USB 4000 fiber optic spectrometer along the depth of the recorded dose distribution using a linear stage (PTW, Freiburg, Germany) which travelled in spatial increments of 1 mm. From all measured spectral data, optical density differences (∆OD) were calculated with Matlab scripts. A relative calibration method of ∆OD to dose was used to calibrate the ∆OD data to dose data.

2.5. Spectroscopic measurements
All spectroscopic samples were taken out of the refrigerator about 45 minutes prior to readout and left to equilibrate at room temperature. Subsequently, the samples were read out by an USB 4000 fiber optic spectrometer using a in-house constructed white led light source. The readout parameters were: 100 ms integration time, 7 nm boxcar width of running average, 50 averages at 632 nm.
3. Results

3.1. Dose sensitivity
The dose sensitivity of the LMD2 dosimeter amounted to $4.375 \times 10^{-3}$ cm$^{-1}$Gy$^{-1}$ (dose rate 400 cGy min$^{-1}$). The dose sensitivity curve shows a linear dependency. The dose sensitivity of the LMD1 dosimeter amounted to $0.8925 \times 10^{-3}$ cm$^{-1}$Gy$^{-1}$ (dose rate 400 cGy min$^{-1}$).

3.2. Dose rate dependence
A clear dose rate dependency is seen for all radiochromic dosimeters investigated in this study. Figure 1 shows that the deviation of the optical dose sensitivity between 50 cGy min$^{-1}$ and 400 cGy min$^{-1}$ of LMD1 type dosimeter reached 74.6% relative to 400 cGy min$^{-1}$ as compared to 33.7% for the LMD2 type dosimeter.

![Figure 1](image1.png)

**Figure 1.** Comparison of dose response for LMD2 dosimeter a and LMD1 dosimeter c irradiated at different dose rates (50 cGy min$^{-1}$ (full line), 100 cGy min$^{-1}$ (dashed line), 200 cGy min$^{-1}$ (dot-dashed line), 400 cGy min$^{-1}$ (dotted line)). Plot b and d show the dose sensitivity as function of dose rate relative to 400 cGy min$^{-1}$. An exponential fit is shown as a full line.

3.3. Depth dose profiles
In figure 2a, depth dose profiles of 6 MeV electron beams are plotted all scaled to the dose maximum measured at 400 cGy min$^{-1}$. In figure 2c all depth dose profiles are individually scaled to the maximum and minimum dose demonstrating the high apparent correspondence between the three curves.
Figure 2. Plot a and c show electron depth dose profiles (6 MeV) measured with the LMD2 type dosimeter at three dose rates: 50 cGy min\(^{-1}\) (red dotted line), 100 cGy min\(^{-1}\) (blue dotted line) and 400 cGy min\(^{-1}\) (green dotted line) compared with an ionisation chamber measured PDD (black full line). The PDD's are scaled with the same normalisation factor for the PDD recorded at 400 cGy min\(^{-1}\) in a. The PDD's are scaled with their individual normalization factor in c. Plot b and d show a simulation of the influence of measured dose rate dependence upon the depth dose profile (dashed line) compared to a PDD recorded with an ionisation chamber (full line).

Additionally, in figure 2b and 2d simulated depth dose profiles are shown as calculated from an ionization chamber measured depth dose distribution and based on the dose rate dependency for electrons.

4. Discussion

4.1. Dose sensitivity

The maximum optical dose sensitivity for LMD2 was 4.375 x 10\(^{-3}\) cm\(^{-1}\)Gy\(^{-1}\) (dose rate 400 cGy min\(^{-1}\)) which is 5% lower than the value of the leucodye dosimeter proposed by Jordan et al. (4.6 x 10\(^{-3}\) cm\(^{-1}\)Gy\(^{-1}\)) [2]. In order to compare the characteristics of the LMD2 dosimeter, the leucodye dosimeter proposed by Jordan et al. (LMD1) was fabricated according an identical fabrication procedure as described in their publication [2]. The maximum optical dose sensitivity for LMD1 amounted to 1.031 x 10\(^{-3}\) cm\(^{-1}\)Gy\(^{-1}\) (dose rate 400 cGy min\(^{-1}\)) which is four times lower than the value reported by Jordan et al. [2]. An explanation for the difference in dose sensitivity was not found. A significant increase of the background color is seen during the first hour post fabrication although the dosimeter was stored in a refrigerator at 4°C. Attempts to reduces this background color by fabricating the gel in a dark room have not been successful.

4.2. Dose rate dependence

Both LMD1 type and LMD2 type dosimeters are shown to be strongly dependent on the dose rate. To illustrate the effect of the dose rate dependency on a dose distribution, a PDD of a 6 MeV electron beam was recorded with a LMD2 type dosimeter irradiated at different dose rates. It is clear that lower dose rate regimes result in higher dose sensitivity (figure 2a). However when all PDD's are rescaled to their dose maximum and minimum (figure 2b) no significant deviation from the ionization chamber measurement is found.

The dose rate dependency effects were also simulated on a depth dose distribution measured with an ionization chamber. The maximum deviation between the dose rate dependent and independent PDD amounts to 8% of dose maximum between 20 mm and 30 mm depth (figure 2d.). However a positional shift of 0.9 mm results in an almost complete correspondence of both curves between dose maximum and 5 cm depth indicating that this method of identifying dose rate dependency is very sensitive to positional errors.

To extend the previous results, the effects of the dose rate dependent optical dose response was also investigated for photon beams. A 6 MV photon beam depth dose distribution was recorded with the LMD2 type dosimeter. Deviations up to 4.9% relative to the dose maximum are found for the LMD2 type dosimeter (figure 3). A dose rate dependent PDD was also simulated based on the measured dose rate dependency of the LMD2 type dosimeter. The results are consistent with a measured PDD.
Figure 3. Comparison of PDD profiles for LMD2 type dosimeter (dotted plot) versus ionisation chamber measurements (full line) of 6 MV photon beams. The dashed line represents the simulated dose rate dependent PDD curve as calculated from the measured dose rate dependency of the LMD2 type dosimeter.

The predominant chemical that influences the dose rate dependency is believed to be the surfactant. Changing the concentration and type of surfactant (SDS versus Triton X100) results in substantial differences in dose rate dependency. A change in gelatin concentrations however does not avoid a dose rate dependent response.

The large number of chemical components and the chemical heterogeneity of the micelles in the LMD type dosimeters results in a high complexity of radiation induced reaction kinetics which cannot be completely disentangled at this moment.

5. Conclusions
The increasing need for dedicated real 3D quality assurance tools in modern radiation therapy is the main motivation behind the development of accurate and user-friendly 3D dosimeters. Recently a novel 3D radiochromic gel dosimeter was introduced which uses micelles to dissolve a leucodye in a gelatin matrix. Experimental results show that the previously introduced micelle dosimeter was found to be significantly dose rate dependent. A novel composition of 3D radiochromic dosimeter is proposed composed of gelatin, sodium dodecyl sulphate, chloroform, trichloroacetic acid and leucomalachite green. The novel gel dosimeter formulation exhibits comparable radio-physical properties and a reduced dose rate dependency, however it is still unsatisfactory for clinical radiation therapy dose verifications. Future research should focus on minimizing the dose rate dependency through physic-chemical analysis of the dosimetry systems. A robust calibration method for ‘absolute’ dosimetry is under development.

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