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Dose rate effects in radiochromic leuco crystal violet dosimeters

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Abstract. Radiochromic dosimeters based on leuco crystal violet (LCV) in aqueous solutions and gelatin hydrogels were evaluated for dose and dose rate performance. Optical transmission measurements were performed on samples in custom 10 cm long cuvettes in order to have measurable signals below 10 Gy. Standard gel formulations with 0.7 mM concentration of anionic surfactant sodium dodecyl sulfate (SDS) had a dose rate effect of increasing sensitivity with increasing dose rate of nearly 15% from 1 to 10 Gy/min at 20 Gy. Solutions of LCV and trichloracetic acid also showed similar dose rate effects. Adding LCV increased dose rate effect. Adding SDS, lowered sensitivity but maintained dose rate effect. Adding 0.4% gelatin also lowered sensitivity but reversed dose rate effect. For gels, lowering the pH reduced both sensitivity and dose rate effect. These trends are consistent with LCV solubility contributing to dose rate effects. These data indicate there are several factors related to dose rate effects in leuco crystal violet dosimeters and formulations designed to maximize sensitivity may not minimize dose rate effects.

1. Introduction

Radiochromic gelatin hydrogels with leuco dyes are one class of 3D dosimeter with several advantages for optical computed tomography (CT) readout of samples greater than 10 cm in diameter. These advantages include: low cost and non-toxic chemicals, easy preparation in open beaker environment, low scatter, low initial colour, low or very low diffusion rates. Initial results indicated gelatin hydrogels with leuco crystal violet (LCV) trichloroacetic acid (TCAA) and non-ionic surfactant Triton X100 was a promising alternate to ferrous-xylenol orange gels for larger volumes because of higher initial transmission and more stable dose image [1]. However, further experiments indicated dose rate effects which would limit conditions for accurate 3D dosimetry [2]. Further experiments in-house found a $\sim 10\%$ fading effect that in certain conditions could be interpreted as an effective dose rate effect. Other groups have found no dose rate effect for this initial formulation [3-5]. Recent large volume samples are measuring a dose rate effect of nearly +15% for increasing dose rate from 1 to 10 Gy/min. Why dose rate effects were not observed previously but are present in recent data is an ongoing investigation. Substituting for nonionic Triton X100 with anionic surfactant sodium dodecyl sulfate (SDS) at 0.7 mM concentration results in a low scatter, non-diffusing, 3D, radiochromic dosimeter [6]. This standard formulation: provides better agreement with known depth doses at short post gelation times than for 3D samples stored at 5 °C for several days. It also has a dose rate effect of near +15% for 12 MeV electrons at 20 Gy from 1 to 10 Gy/min and under-responds in low dose regions of arc delivery treatments. While, 12 MeV electron depth dose and repeat irradiations of small, 1 cm, cuvettes both show linear dose responses to >40 Gy, repeat irradiations of larger volume samples and 3D reconstructions reveal dose

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responses dependent on dose rates of previous and current deliveries. These sequential 3D irradiations are a sensitive test of combined effects of dose and dose rate in the same volumes as intended unknowns.

Because of the mixed results in the literature and current problems with this SDS formulation a set of experiments were conducted to re-examine dose rate effects. These include, examining solutions and gel formulations in custom 10 cm long by 2.2 cm diameter polymethyl methacrylate (PMMA) cuvettes. The longer cuvettes allow accurate transmission measurements for lower doses and the larger volumes may be more representative of results in the bulk of 3D dosimeters away from the vessel walls.

2. Methods

Leuco crystal violet (LCV), trichloroacetic acid (TCAA), sodium dodecyl sulfate (SDS) and gelatin (porcine, type A, 300 bloom) were purchased from Sigma-Aldrich. The order in which samples are added can produce different results. In general, the water or gel solutions are lowered in pH before adding SDS or LCV. This minimizes initial colour change. Dispersing SDS before addition of LCV results in lower scatter samples. Concentrations similar to current optimum gel formulation (5% gelatin+32 mM TCAA +1 mM LCV +0.7 mM SDS), were starting values.

The samples were usually prepared a few hours prior to irradiation and maintained at 20 °C throughout experiments. Photochromic tests indicated no measurable change in transmission for the light exposures of this study. Custom 10 cm long PMMA cuvettes were prepared by cutting fill/cleaning slots along lengths of 2.5 cm OD plastic tubing and welding 0.15 cm thick PMMA plates to the ends. After filling, samples were sealed with clear 3M, Scotch[™] tape. Transmission measurements were performed with a Hitachi-Perkin Elmer model 204 absorption spectrometer at 590 nm with 1 mm slits and a water reference. Linear attenuation coefficients were calculated for direct comparisons with optical CT reconstructions. Irradiations were performed with a Varian 2100 IX series linear accelerator; 6 MV, 6 Gy/min, jaw size 20x20 cm or 12 MeV at 1 or 10 Gy/min, 4x4 or 20x20 cm cutouts. For dose rate experiments, cuvettes were irradiated as two groups in a water tank with cuvette axes at 2.8 cm depth. Irradiation and readout required 20 minutes between subsequent dose points.

3. Results

Even and odd numbered samples were irradiated at 10 and 1 Gy/min dose rates respectively. In figure 1, the stock solution was 1 mM LCV + 25 mM TCAA and S0 and S1 are same solution irradiated at 10 vs 1 Gy/min. The plot demonstrates a dose rate effect which increases with dose and has greater sensitivity for higher dose rate similar to the standard gel formulation. S10 and S11 contain stock solution + 0.7 mM SDS. This concentration of SDS lowered dose sensitivity by 50% and maintains the trend in dose rate effect. Note, 0.7 mM is below the critical micelle concentration of SDS in water. Adding 0.4% gelatin to stock solution (S40, S41) reduced sensitivity by $\sim 65\%$ but reversed the trend in dose rate effect. Specifically, the lower dose rate is $\sim 5-10\%$ more sensitive. All three samples are sublinear with dose response. S50 combines SDS with gelatin, resulting in an intermediate sensitivity but increased linearity in dose response. No sample S51 was available for comparison. These results indicate dose rate effects are dose dependent and have multiple causes.

In figure 2, the dose range was increased to 60 Gy. S4 and S5 curves show solution sensitivity at 30 Gy increased 30% by increasing LCV concentration from 1 to 2 mM, see figure 1 S0 and S1. Also the dose rate effect increased with more LCV in solution. Sublinear dose response was similar for 1 and 2 mM solutions. Increasing the TCAA concentration to 35 mM, lowered the dose sensitivity and reversed the dose rate effect below 30 Gy compared to S0 and S1. At higher doses, the dose rate effect was minimal. These results indicate solutions with LCV concentration <1mM and TCAA concentration >25 mM may have smaller dose rate effects at doses <30 Gy. In contrast, a gel formulation, S90 (4% gelatin + 1 mM LCV + 0.7 mM SDS + 25 mM TCAA), has an intermediate dose sensitivity which is linear to >60 Gy.

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Figure 1. Plot of solution attenuation coefficient vs dose in 10 cm cuvettes with repeat irradiations. Solid/open markers at 10/1 Gy/min respectively. Reference, 0 Gy sample.



Figure 2. Plot of solution and gel attenuation coefficient vs dose in 10 cm cuvettes with repeat irradiations. Solid/open markers at 10/1 Gy/min respectively. Reference, 0 Gy sample.

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Figure 3. Plot of gel attenuation coefficient vs dose in 10 cm cuvettes with repeat irradiations. Solid/open markers at 10/1 Gy/min respectively. Reference, 0 Gy sample, dotted line, linear fit to S100 gel, solid line drawn as a visual guide.

In figure 3, the effects of increasing the TCAA concentration from 25 to 30 and 34 mM in 4% gelatin gel is shown. Note, samples S92 and S93 are from same batch as S90 but two days older at irradiation. Essentially, the sensitivity and dose rate effect decrease with increasing TCAA concentration. This trend suggests that LCV may not be completely dissolved at TCAA concentration of 25 mM in 4% gelatin or 32 mM in 5% gelatin (common formulations). Also shown, is another formulation, T0 and T1 where the gelatin solution is pretreated with 1 mM hydrogen peroxide. This step increases the initial gel dose sensitivity threefold. However, after several days of storage at 5 C the sensitivity decreases to similar levels found without this step. Note the dose rate effect for this same day sample is reversed. This data supports the potential complexity of gelatin hydrogel dosimeters.

4. Conclusion

Comparisons of solution and gel dosimeters for same chemistry provides additional information for development of hydrogel dosimeters. Ten cm long cuvettes provide more accurate measurement of dose response below 10 Gy. However, direct comparison with performance in large volume (10 cm diameter or larger vessels) is necessary before relevance of data can be determined. Other similar, in-house, experiments, indicate, larger dose rate effects can be measured in large samples with 3D optical CT readout. This suggests cuvettes, 10 cm long with 2.2 cm inner diameter are not sufficient to reproduce 3D gel response. The data suggests that focus on maximizing dose sensitivity may result in formulations with measurable dose rate effects.

5. References

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