Non-diffusing radiochromic micelle gel

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Abstract. The addition of Laponite, a synthetic clay nanoparticle material to radiochromic
leuco Malachite Green micelle hydrogel eliminates diffusion of the cationic dye by
electrostatic binding. The clay nanoparticles also increased dose sensitivity ten-fold relative to
the parent gel formulation. This material is a suitable 3D water equivalent dosimeter with
optical CT readout.

1. Introduction

Transparent, radiochromic hydrogels are ideal candidates for tissue or water-equivalent radiation
dosimetry with optical readout. Materials with minimal scatter can be efficiently scanned by both broad-beam and single-ray optical CT scanner geometries. Also, low scatter materials are preferred for scanning larger volumes, such as cylinders up to 30 cm in diameter. Radiochromic micelle gelatin hydrogels have been reported for both leuco malachite green (LMG) [1] and leuco crystal violet [2]. The respective diffusion coefficients of these gels are 3 and 25 times lower than the reference ferrous xylene orange gel (FX). While these gels are promising for quantitative 3D dosimetry, the current formulations have 30 and 10 times lower dose sensitivities relative to typical FX gel formulations.

Clays are known to provide binding surfaces for solvents and solutes. In particular, clays are under investigation as low cost materials for trapping dyes in effluent from industrial textile production. Laponite is a synthetic nanoparticle clay which has several chemical properties which may be useful for improving the performance of radiochromic hydrogels. Laponite, binds cationic dyes through an electrostatic interaction [3], and binds to collagen in gelatin hydrogel [4]. At higher concentrations it directly forms transparent, colourless hydrogels. This preliminary investigation reports that introducing a small amount of Laponite into LMG micelle gel eliminates the diffusion of malachite green formed after irradiation of the hydrogel.

2. Methods

The gel formulation was: 4% gelatin, 0.1% Laponite, 15 mM trichloroacetic acid, 7 mM Triton X-100 and 0.3 mM LMG. The gelatin (porcine, 300 bloom, Sigma) was slowly added to water (at 22 C, deionized Millipore) and then stirred in a water bath at 40 C. Similarly, the Laponite RD (sample from Southern Clay Products, Tx USA) was sprinkled on water at 22 C and stirred to form a clear colourless solution. An aqueous solution containing, Triton X-100 (Sigma-Aldrich reagent grade), trichloroacetic acid (Mallinkrodt, Cat No. UN1839) and LMG (Aldrich reagent grade) was stirred at 22 C to form a transparent, pale green solution. The Laponite solution was added slowly to the warm gelatin solution and mixing continued for two hours at 40 C. After lowering the gelatin solution to
30°C the surfactant solution was slowly added while stirring and poured into 1 cm plastic cuvettes. The samples were stored at 4°C for gelation. Samples were irradiated under conditions of full scatter with 4 or 6 MV x-ray beams from medical linear accelerators at dose rates near 4 Gy min⁻¹. For dose response measurements, transmission at 633 nm was measured between irradiations, with about 10 minutes between points. For diffusion studies, half of a cuvette was irradiated to 5 Gy and a sequence of transmission images at 590 nm were recorded at 22 C.

3. Results
The dose sensitivity of this gel increased with concentration of LMG, Laponite and gelatin. Sensitivity was also related to concentrations of surfactant and AC13. Increasing Laponite concentration from 0.1 to 0.2% by mass increased both scatter and dose sensitivity. In figure 1, transmission profiles recorded 0.5 and 3.5 hours post irradiation are shown. Except, for an offset due to auto-oxidation the two profiles are essentially the same. This demonstrates that the MG⁺ dye that is formed is immobilized when Laponite is present. In figure 2, the dose response for this formulation is presented. The dose sensitivity is around 25% of typical FX gel formulations. All formulations investigated demonstrated a linear doses response with development times less than one minute and no fractionation effects.

![Transmission Profiles](image)

Figure 1. Transmission profiles for LMG-micelle-Laponite gel in half irradiated cuvette at 0.5 (-) and 3.5 (■) hours post irradiation, air reference.
4. Summary
Laponite RD clay nanoparticles when added to radiochromic LMG micelle gels eliminate diffusion and increase the dose sensitivity roughly ten fold. The Laponite also causes a moderate increase of optical scatter in the gelatin gels. Laponite may be a general chemical approach to elimination of diffusion in radiochromic gels that contain cationic dyes.

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