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Review of recent advances in radiochromic materials for 3D dosimetry

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Abstract. Recent papers concerning radiochromic films, plastics and hydrogels for 3D dosimetry are summarized. The utility of Presage™, a radiochromic plastic, with optical CT readout was demonstrated for the following applications: motion and gated treatment delivery, commissioning of small fields for radiosurgery, ¹⁹²Ir high dose rate brachytherapy source commissioning and as a 3D insert for IMRT credentialing tests with Radiological Physics Centre (RPC) phantoms. Preliminary performance for characterizing microbeams from a synchrotron with optic projection tomography readout demonstrated resolution of an 83 micron diameter beam. Hydrogel chemistries based on nonionic micelles for leuco malachite green and leuco crystal violet demonstrated that low diffusion gels can be designed by choosing product dyes that are poorly soluble in water and tend to remain in the micelles. Turnbull blue chemistry has been successfully adapted to form a non-diffusing gel as well. The performance of ferrous xylenol orange hydrogel layers doped with boron to form neutron dosimeters demonstrated another practical application. Polymerization hydrogels are alternate materials that can be read with optical CT scanners. High dose gradient applications in brachytherapy with ⁹⁰Sr/⁹⁰Y sources and proton dosimetry are presented for comparison.

1. Introduction

This review provides a summary of works published since the 5th International Conference on Radiotherapy Gel Dosimetry, DOSGEL 2008. The material draws primarily on search results within the PUBMED and Web of Science databases using the keywords; 3D, three-dimensional, dosimetry and radiochromic. Polymerization gels have been treated as a separate topic since many gel dosimetry researchers have been focused on these materials and chemistry and readout reviews were presented earlier in this conference. However, several polymer gel papers that involve optical CT readout have been included here as well for convenient comparison. Radiochromic materials are generally read by optical transmission techniques. In particular optical computed tomography (CT) scanners are the primary technology for 3D scanning. There are several scanner geometries and they are broadly classed as single ray – single detector or broad beam (fan beam, cone beam and collimated beam) – array detectors. The single ray - single detector geometries are least sensitive to scatter in contrast to high acceptance angle cone beam systems being the most sensitive. For this reason, the quality of the reconstructed 3D dose distribution is intimately connected to both the optical performance of the dosimeter and readout scanner. Advances in scanner design, data processing, algorithms for reconstruction and materials development provide steady improvements in quantitative tissue/water

equivalent 3D dosimetry. Fast scanners make 3D dosimetry more practical to perform in a clinical setting and allow gels such as ferrous-xylenol orange to accurately record high dose gradients generated with megavoltage radiation beams. Materials research to develop higher performance materials concentrates on: low diffusion chemistries, rapid dose response, adequate sensitivity for the typical two Gray radiotherapy treatments and low scatter in order to achieve the highest spatial resolution and dynamic range.

There are currently three materials for radiochromic 3D dosimetry; film (EBT2, International Specialty Products), gels and plastics (Presage™, Heuris Pharma LLC). By default EBT2 film is becoming the film standard for radiotherapy QA measurements and a dedicated session is included in this conference. In this review, there is overlap with many of the other presentations because most of the current polymer gel formulations can be read in 3D with optical CT, magnetic resonance imaging (MRI) and x-ray CT. The issue of scatter contamination in transmission profiles is generally addressed by choosing scanner geometries with low acceptance angles such as single ray- single detector or fan-beam with linear array detectors. However, substantial progress in scatter rejection for cone beam CT provides stimulus for continued scanner development of all geometries. As well, the optical scanners can easily be adapted for film densitometry providing added value.

In order to compare different dosimetry systems for a specific application there are a few basic parameters required in order to determine which materials and readout scanners are adequate. Irradiation criteria will include: dose range, dose rates, energy of radiation beam, spatial resolution and volume of interest. The dose sensitivity, which is expressed as the change in optical linear attenuation coefficient per Gray (units $\text{cm}^{-1} \text{Gy}^{-1}$) can quickly eliminate many materials if the maximum dose is around 2 Gy. A very useful metric is the dose resolution of a system [1]. It provides a measure of the minimum dose that can be measured with a specified uncertainty for a given spatial resolution. For example, a comparison at a fixed spatial resolution such as $1 \times 1 \times 1$ mm voxel size quickly ranks the relative performance of complete systems. However, it does not identify the limiting factors which may be related to the dosimeter or the readout mechanism. Temperature needs to be controlled in order to conduct reproducible chemical reactions and the thermal history of the material may also be a factor. Most materials that have sufficient dose sensitivity are likely also sensitive to light and heat. Drifts in absorption coefficients are generally due to auto-oxidation rates. Ferrous xylenol orange gels continue to darken whereas materials based on leucodyes may darken or bleach depending on the specific chemistry. The rate at which the radiochromic reaction proceeds can have a negative effect. For example, if diffusion of radiation induced chromophores is an issue then the reaction needs to be fast so that 3D scanning can be completed prior to degradation of the dose distribution gradients. Fast reactions are very useful from a practical perspective as well. Some systems now allow for 3D scanning and reconstruction to be completed in less than 30 minutes post irradiation. Dose response data is important for development of radiation sensitive materials. Common issues include: linearity, low dose response, dose rate and fractionation effects. Often there are measureable differences with sample volume that make usage of cuvettes inadequate for calibration of larger volumes. Both the scatter and absorption coefficients are important for estimation of material performance. For radiochromic reactions, transparent and initially colourless materials are preferred in order to maximize dynamic range and spatial resolution. Material composition is required for calculation of electron density and mass density for scaling to water or tissue equivalent dimensions.

A recent review by Doran has provided a concise history of optical CT and dosimeters for 3D dosimetry [2].

2. Dosimeter developments and applications

The following papers contain a mix of dosimeter material results as well as instrumental innovations that were required to attain these signal to noise ratios. The applications papers also contain information related to improvements in materials, instruments and data analysis.

2.1 Radiochromic film

Silver based film is being rapidly phased out due to increasing costs of the dwindling commercial supply. The major film manufacturers have evolved in response to the near universal adaptation of solid state array detectors for image detection. Film dosimetry in practice is becoming limited to the set of radiochromic films available from International Specialty Products. The particular product for megavoltage radiotherapy dosimetry is referred to as EBT2. There are many recent publications concerning evaluation of this product. A detailed uncertainty analysis for radiochromic film scanned with a flatbed document scanner, demonstrated an uncertainty of 0.4% is achievable [3]. As the quality of 3D dose distributions improves, analogous uncertainty calculations will be required when comparing to equivalent planes with film measurements. EBT film is no longer commercially available and the early batches of its successor EBT2 have demonstrated some variability in energy response below 100 keV [4]. EBT film has been the standard 2D dosimeter for comparison with selected planes of 3D dosimeter results. This implies that researchers using this material need to be proficient in its use in order to attain quality data for comparisons.

A report of stacking 8 EBT film strips to record near surface doses, at depths ranging from 0.012 to 0.18 cm, reported agreement with parallel plate ion chamber and thin TLD measurements [5].

2.2 Radiochromic plastic: Presage™

A very promising material for practical 3D dosimetry is the radiochromic plastic, Presage™. Leucodyes and halogenated hydrocarbons are dissolved in polyurethane to form a robust dosimeter. The material does not exhibit diffusion and can be formulated to cover any dose range of relevance to radiotherapy. It is anticipated that versions of this material will become one of the common 3D dosimeters for routine clinical usage. Most results are reported with leucomalachite green (LMG) formulations and readout near 633 nm. This wavelength is very convenient since EBT films also have their main absorption peak at 639 nm. This allows optical CT scanners optimized for Presage™ to also read EBT film. Since early Presage™ formulations contained halocarbons initiator content in the range of 5 to 20% by mass the low energy response is expected to be larger than for water. Also the carbon to oxygen ratio is much greater than in hydrogels leading to an effective atomic number higher than for water. These effects were quantified in a Monte Carlo calculation comparing PAGAT polymer gel and a specific formulation of Presage™ [6]. The calculations reported the differences in depth doses for 6 and 15 MV photon beams relative to water for both dosimeters. Results demonstrated that PAGAT was nearly water equivalent and the Presage relative depth dose increased up to 7% at 30 cm depth. Recent formulations are reported to be more water equivalent. Other researchers have prepared Presage™ based on published papers and patent and have obtained similar results. The sensitivity and long term stability as a function of LMG and initiator content was recently reported by Mostaar et al [7].

Sakhalkar et al have reported on the performance of Presage™ for the specific application of 3D dosimetry within the IMRT head and neck phantom from the Radiological Physics Centre (RPC) [8-10]. The dosimetry insert contains TLD's and orthogonal EBT films. Substituting a Presage cylinder for the insert allowed for 3D dosimetry within the pocket. Multislice scanning with a commercial laser CT scanner (OCTOPUS 5x) revealed better agreement with EBT film results than with the treatment planning calculation. A set of four samples were used to demonstrate interdosimeter reproducibility to within 2% for dose difference and 2 mm distance to agreement relative to the average response. This particular formulation demonstrated a postirradiation increase in absorption that scaled uniformly throughout the dose distribution. The successful implementation of Presage™ into the RPC credentialing process will provide high profile application of 3D dosimetry.

The use of Presage™ to verify gated radiotherapy delivery has been reported by Brady et al [11]. This dosimetry problem has been recognized as difficult for point dosimeters and 3D dosimeters have provided convincing data for the utility of 3D dosimeters. A CCD camera with telecentric lens was used to capture the transmission images. Improvements to this broad beam optical CT scanner are also reported to increase signal to noise levels 3-fold over previous reports.

Full 3D dosimeters have been used for small field dosimetry. They have the advantage of being both the phantom and detector, which simplifies the irradiation process. Because Presage™ has no reported diffusion it is anticipated to perform well for commissioning small fields for radiosurgery. Clift et al presented depth doses, profiles and output factors for Presage scanned with CCD camera with telecentric lens and laser CT (OCTOPUS 5x) which agreed with EBT film and previous commissioning data measurements [12]. The results demonstrate the efficiency of 3D dosimetry for commissioning small fields.

Proton dosimetry is yet another dosimetry application that involves high dose gradients. Because of the high LET of the protons near the end of their range, recombination of radicals has made quantitative chemical dosimetry inadequate for this particular problem. An under response has generally been observed with hydrogels (exception BANG3-Pro gel, see below) and previous Presage™ formulations. This issue has been re-examined and an under-response of 20% was observed for the Bragg peak [13]. Microbeam irradiation with synchrotrons has generated exciting radiobiology results that are key for understanding the mechanisms of grid therapy. These beams are typically less than 100 microns in diameter and require much higher spatial resolution than is required for IMRT type distributions. Only dosimeter materials that do not exhibit diffusion are relevant for measuring these high gradients. Early results for Presage™ with a high resolution optical projection tomography system revealed dose lines less than 80 micron widths could be resolved [14]. Brachytherapy is another common dosimetry problem in which high dose gradients are present. The utility of Presage™ to accurately record the distribution from a high dose rate ^{192}Ir source were presented by Wai et al [15].

2.3 Micelle hydrogels

The addition of surfactants above a threshold concentration allow micelles to form. In hydrogels, the interior of the micelles provides a nonpolar chemical environment in which nonpolar reactants can dissolve. This innovation allows the opportunity to design radiation sensitive reactions based on nonpolar chemistry within hydrogels [16]. These reactions could be polymerization or radiochromic reactions. The initial results have focused on radiochromic systems. Non-ionic surfactants such as Triton X100 with LMG and trichloroacetic acid form transparent gelatin hydrogels. Following a 5 Gy preirradiation, these gels demonstrate a linear response. The linearity is quantified by obtaining a direct correlation between a 12 MeV electron beam depth dose and the corresponding attenuation coefficient as determined by optical CT. This initial formulation had a dose sensitivity of $0.004 \text{ cm}^{-1} \text{ Gy}^{-1}$ which is approximately 3% of typical FX formulations. It also had a diffusion coefficient approximately 3 times lower than FX gels. While this gel demonstrated feasibility of micelles for developing new radiation sensitive gels it has limited value for clinical dosimetry based on its low sensitivity. A subsequent study with leuco crystal violet (LCV) provided a much improved material. The sensitivity was doubled and the diffusion was 10 times less than the LMG version [17]. The LCV gel demonstrated a linear response with no requirement for preirradiation. These gels are easy to prepare and provide a convenient alternative to FX gels provided higher doses can be delivered. One application where they can be a valid alternative to FX gels is for commissioning of small fields [18].

2.4 Radiochromic Turnbull Blue gel

An alternative radiochromic chemistry is based on the production of Turnbull blue $(\text{Fe(II)Fe(III)CN}_6)^+$. A new hydrogel based on phytigel and ferric cyanide chemistry had been reported [19]. This material has an absorption peak around 690 nm, very low diffusion rate and dose sensitivity around $0.005 \text{ cm}^{-1} \text{ Gy}^{-1}$. This sensitivity is similar to the LMG micelle gel but with the advantage of no diffusion. Samples demonstrate sensitivity to ambient lighting and a measureable auto-oxidation rate which is non uncommon for radiation sensitive materials.

2.5 Ferrous xylenol orange gels

Ferrous xylenol orange gelatin gel continues to be a useful material in spite of the diffusion rate. In cases where irradiation times are short, gradients modest (typical IMRT type gradients, <10% per mm) and scan times are short these gels provide adequate results. Care is required concerning thermal history. Depending on wavelength of readout a preirradiation dose may be required in order to establish a subsequent linear dose response. The recent innovations have concentrated on the addition of boron to the gels in order to perform neutron dosimetry [20-23]. Calculations reporting the water equivalence of FX and several formulations of polymerization gels have also been reported [24,25].

2.6 Polymer hydrogels

A considerable fraction of 3D hydrogel dosimetry research has concentrated on polymerization gels in which monomers undergo free radical induced polymerization. The literature has been summarized in a review article authored by many of the pioneers in this area [26]. The commercial versions are referred to as BANG™ gels. Several formulations are available, optimized for specific dosimetry problems. The utility of BANG gel with readout by high resolution optical CT has been reported for high gradient applications [27,28]. Verification of gated treatment delivery has also been reported [29]. In the case of proton dosimetry, a specific formulation has successfully recorded the Bragg peak, demonstrating equivalence to ion chamber depth dose curves [30].

3 Optical CT scanner design

Incremental improvements to scanner designs for laser CT and broad beam systems with CCD cameras have been included in previous references related to application studies. Another geometry exploring the performance of free space scanning has been reported. Elimination of the aquarium and refractive index matching liquid would simplify scanning if sufficient projection data could be obtained. The work compares polymer gel dose distributions with the prototype optical scanner and MRI reporting similar results for specific geometries [31]. Further work may be required to determine if this approach is useful for scanning complete sample volumes.

4 Summary

Radiochromic materials development continues to be an active area of research. The development of several strategies to produce low or zero diffusion materials allows many high dose gradient applications to be effectively measured with radiochromic systems providing an alternative to polymerization gels. Optical CT continues to improve as experimenters address issues with specific geometries and explore alternate geometries. Several scanner geometries now report full 3D data scans in less than 10 minutes which minimizes the impact of diffusion for IMRT type gradients.

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