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PRESAGE® as a solid 3-D radiation dosimeter: A review article

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ABSTRACT

Radiation oncology has been rapidly improved by the application of new equipment and techniques. With the advent of new complex and precise radiotherapy techniques such as intensity modulated radiotherapy, stereotactic radiosurgery, and volumetric modulated arc therapy, the demand for an accurate and feasible three-dimensional (3-D) dosimetry system has increased. The most important features of a 3-D dosimeter, apart from being precise, accurate and reproducible, include also its low cost, feasibility, and availability. In 2004 a new generation of solid plastic dosimeters which demonstrate a radiochromic response to ionizing radiation was introduced. PRESAGE® plastic dosimeter lacks the limitations of previous Ferric and polymer plastic 3-D dosimeters such as diffusion, sensitivity to oxygen, fabrication problems, scanning and read out challenges. In this decade, a large number of efforts have been carried out to enhance PRESAGE® structure and scanning methods. This article attempts to review and reflect on the results of these investigations.

1. Introduction

Radiation oncology has been rapidly improved by application of new equipment and techniques. These techniques have been established based on delivering high radiation doses inside small treatment volumes with variable dose rates and leaf movement that ultimately lead to creation of homogeneous and desirable dose distribution in the target with minimal dose to normal tissues (Gautam, 2014; International Commission on Radiation Units and Measurements (ICRU), 2010). Although, conventional dosimeters, such as ionization chambers, thermoluminescent detector (TLD), and diodes can measure absolute dose with high precision; due to their large sensitive volume, the measurements in a small field with high dose gradient is challenging (Das et al., 2008a; McKerracher and Thwaites, 1999; Wuerfel, 2013). At the moment, treatment verification in intensity modulated radiotherapy (IMRT) techniques have been implemented by point dosimeters such as diamond, ionization chamber and the two-dimensional dosimeters such as film, two dimensional-array detectors; however, three-dimensional (3-D) verification has had some challenges (Das et al., 2008b; Ezzell et al., 2009; Low et al., 2011). The history of gel dosimeters as a kind of chemical 3D dosimeters dates back to 1950, when Day and Stein reported a color change in Folin's Phenol due to radiation exposures (Doran et al., 2009; McJury et al., 2000). In 1958, Hoecker and Watkins introduced the polymerization of monomers induced by ionizing radiation as a process for dosimetry (Hoecker and Watkins, 1958).

According to chemical mechanism, gel dosimeters have been divided to three main groups: Ferric dosimeters, polymer and radiochromic gel dosimeters. Ferric gels were developed by Gore and Kang (1984) (Gore et al., 1996). The radiation sensitive part is Ferrous ion (Fe^{2+}). Upon irradiation, a water free radical oxidizes the ferrous ions (Fe2+) to ferric ions (Fe3+) (Balcom et al., 1995; Podgorsak and Schreiner, 1992). The ionic radius and magnetic moments of ferrous and ferric ions are so different that it results in the formation of different bonds in terms of strength and solidarity with water molecules; this characteristic is responsible for the changes in spin-lattice parameter of relaxation time (T_1) of the protons in water molecules. Therefore, the spin-lattice relaxation rate $R1 = (1/T_1)$ can be illustrated as a dose map using magnetic resonance imaging (MRI) scanning. However, MRI is a complex method widely used for imaging, but quantitative analysis with MRI is affected by a lot of factors that restrict reproducibility of results, such as RF coil tuning, physical position

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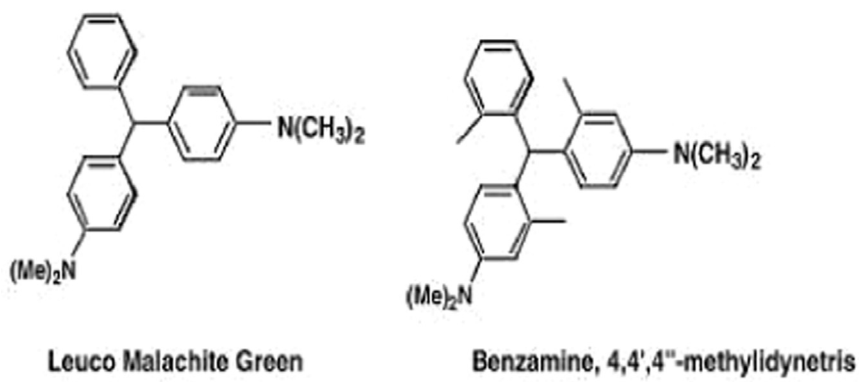

Fig. 1. Chemical formula of the radiochromic response. Reproduced with permission from Journal of Physics: Conference Series (Adamovics and Maryanski, 2004).

within the coil, coil loading, imaging slice orientation and room temperature (Baldock et al., 2010; McJury et al., 2000). Another kind of ferric gel dosimeter with Xylenol Orange dye (FeXO) as radiochromic agent can be classified in ferric or radiochromic 3D dosimeters. FeXO dosimeter is a ferrous sulfate aqueous solution that, when irradiated or subject to heat, oxygen and light, oxidizes the ferrous ions (Fe^{2+}) to ferric ions (Fe^{3+}) and this new type of ion together with xylenol orange form the Fe3+ xylenol complex (Babic et al., 2008). However, diffusion problem is a challenge in low viscos liquid base of ferric gel (Baldock et al., 2001).

The effect of radiation on degeneration of a polymer were first proposed by Alexander (1954) and then Hoecker investigated polymer gel dosimeters (1958) (Alexander et al., 1954; Baldock et al., 2010). The sensitive part of polymer gels generally is Acrylamide or Methacrylic monomers, which are converted to polymers following irradiation. Polymerization process begins when free radicals are created in a gel matrix by radiation. So, the excitability of water molecules changes by the polymerization in sensitive gel, hence the spin-spin relaxation time of water molecules is changed, therefore the dose map in the gel can be obtained using by T_2-weighted MRI scan (Baldock et al., 2010; Crescenti et al., 2007; McJury et al., 2000).

In radiochromic dosimeters, upon irradiation, radiolysis produces free radicals and oxidizing dye species and converts it to a color agent. In 2004, Adamovics proposed another type of radiochromic solid base dosimeter and it was patented named "PRESAGE®" (Adamovics, 2006). It shows a radiochromic response upon ionizing radiation (Adamovics et al., 2004). Some special inherent properties of this radiochromic dosimeter make it an attractive candidate for 3-D dosimetry. The purpose of this study is to review the previous efforts on polyurethane base radiochromic gel dosimeter as named PRESAGE® that during past decade has been progressing in two parallel fields, improvement of PRESAGE® and scanning method.

1.1. Advantages and disadvantages of PRESAGE®

PRESAGE® is a radiochromic solid dosimeter; therefore, it can be fabricated in any desired shape without any container, which is very important in optical scanning of PRESAGE®. One of the disadvantages of ferric is diffusion of radiated part in matrix gel that causes varying of spatial resolution of dosimeter over time; however, PRESAGE® has solid matrix. In megavoltage energies, PRESAGE® is tissue equivalent and its response is independent of the room temperature, dose rates and wide range of energies. Another advantageous characteristic of this plastic dosimeter is that the heat effect on its spontaneous polymerization at temperatures below 80 °C is negligible (Adamovics et al., 2006a, 2006b; Adamovics et al., 2004; Guo et al., 2006b; Farhood et al., 2017).

Recently a deformable PRESAGE® formulation was introduced which exhibits high potential for reusability, with strong post-irradiation optical clearing (Juang et al., 2013; Pierquet et al., 2010a). PRESAGE® dosimeter is not only insensitive to oxygen, but the presence of oxygen during fabrication can improve the sensitivity as well (Alqathami et al., 2015).

2. PRESAGE® structure and method of fabrication

The main component of PRESAGE® dosimeter is polyurethane which has wide applications in medical equipment, construction of coating equipment, adhesives and sealants (Adamovics et al., 2006a, 2006b; Adamovics et al., 2004). There are three characteristics of polyurethanes that make them ideal for radiochromic based dosimetry. It has a solid form, clear texture, and it can be polymerized at a relatively low temperature (< 80 °C), which minimizes undesired thermal oxidation reactions that increase the background of radiochromic response (Adamovics and Maryanski, 2006). However, Adamovics et al. had already tested acrylics, epoxies, polycarbonates, polyesters, polystyrene, polyurethanes, and Polyvinyl-chlorides as base materials of PRESAGE®. Finally, they concluded that polyurethane was more appropriate than the other materials. For other base matrix, they noted that the effective atomic number (Z_{eff}) value of Polyvinylchloride is not the same as tissue. The production of high heat (> 100 °C) during polymerization of acrylics, polycarbonates, polyesters and polystyrene can degrade leucodye in dosimeter. Also, epoxy possesses a low radiation sensitivity (Adamovics et al., 2006a). The chemical formula of Polyurethane commonly used in all studies consists of 20% oxygen, 61% carbon, 9% hydrogen and 10% nitrogen, with $Z_{eff}=6.6$ and density of 1.05 gr/cm (Adamovics and Maryanski, 2004, 2006).

The main radiochromic component of PRESAGE® is a leuco-dye and a free radical initiator (RI). For the radiochromic leuco-dye, a wide range of organic materials can be used (Adamovics et al., 2006b; Adamovics et al., 2004). However, Leuco-Malachite Green (LMG), one of the derivatives of triphenyl methane, is more widely used in the fabrication of the PRESAGE®. In the irradiated part of PRESAGE®, the color change does not occur unless the RI is present in dosimeter formulation. Upon irradiation, halocarbon radiolysis occurs and produces free radicals which oxidize LMG into the malachite green (MG) (Adamovics et al., 2004). Fig. 1 depicts the chemical formula of LMG and MG. Various kinds of organic peroxides, halocarbons, azo, and carbonyl and sulfur-components can be used as a RI. Halocarbons such as chloroform, carbon tetrachloride, and methylene chloride can cause the oxidation of the leuco-dye in water systems (Adamovics et al., 2004; Strukul, 1992).

Fabrication of PRESAGE® consists of two steps, the first step involves mixing of one equivalent of polyols with two equivalents of di-isocyanate. Polyols are a type of alcohol containing multiple hydroxyl groups (OH) with active hydrogen which are used as primary aromatic polymer in the petrochemical industry. Di-isocyanate is a derivative of isocyanate which is a type of organic compounds with the general formula of N = C = O. The reaction of these compounds with polyol converts them to polyurethane. The product is a non-reactive component which can be stored at room temperature. The chemical reaction