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Investigating the potential of polymer gel dosimetry for interventional radiology. Preliminary results.

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Abstract: Complex interventional radiology (IR) procedures contribute an increasing percentage of the overall medical radiation exposure of the population making accurate dosimetry a challenge. Magnetic resonance (MR) based polymer gel dosimetry has been widely employed in complex dosimetric problems in radiotherapy. The aim of this paper is to investigate the feasibility of normoxic gel dosimetry in IR. Dose response, energy dependence and dose rate dependence were investigated in irradiation setups relevant to IR for a particular normoxic gel, based on Methacrylic Acid (MAA) as the monomer and including THPC (Tetrakis-hydroxy-methyl-phosphonium chloride) as antioxidant.. The gel presents a linear dose response beyond a 25 cGy threshold. While no significant energy dependence was observed in the useful range of interventional radiology (80-110kVp) at the low dose level, a linear correlation between gel response and dose rate was observed in the range of dose rates relevant to IR (5-8 cGy/min). These results may suggest that oxygen scavenging of free radicals and polymerization inhibition reactions by hydroquinone have a quantitative effect at gel response at very low dose rates.

Keywords: polymer gel, normoxic, interventional radiology, energy dependence, dose rate dependence, radiation dosimetry, nuclear magnetic resonance, relaxation.

1. Introduction

Interventional radiology (IR) procedures are increasingly being employed as fluoroscopy guided diagnostic and therapeutic interventions, thus contributing a growing proportion of the overall medical radiation exposure of the population (Padovani et al 2005). As a consequence, dosimetric studies in interventional radiology are currently drawing attention (Padovani et al 2005, Vano et al 2005, Kosumen et al 2005, Tsalafoutas et al 2006).

While it is usually sufficient to measure or calculate averaged dosimetric quantities sometimes the need arises for exact dose calculations in a significant volume of tissue. Most IR dosimetric studies use dose area product (DAP) or the equivalent dose to derive a reliable calculation of the total stochastic risk for each procedure. Additionally, deterministic skin effects are addressed by measuring skin entrance dose using TLD arrays, photographic paper and slow radiochromic films or even more elaborate methods like on-line dosimetric software (Vano et al 2005, Kosumen et al 2005,Chu et al 2006). While in most cases it is sufficient to measure or calculate averaged dosimetric quantities for general purpose effective patient dosimetry, sometimes the need arises for exact dose calculations in a specific volume of tissue, especially for IR procedures that are repetitive and/or involve irradiation of sensitive organs or tissues. Radiation dose delivered to sensitive internal organs can only be derived by indirect calculation using either averaged or specific anthropometric and dosimetric data (ICRU Report 74 2005), by Monte Carlo simulations (Bozkurt and Bor 2007). Another approach is to use specialized anthropomorphic phantoms (Meric et al 2005), although this produces dispersed point measurements within the volume of interest corresponding to generic (not specific) anthropometric data. Such methods, however, introduce significant uncertainties due to their indirect nature and implementation variations (Tsapaki 2001).

Direct 3-dimensional (3D) dose distributions of the irradiated volume can only be produced by radiosensitive gel dosimetry. Radiosensitive gels are based on various forms of chemical dosimetry such as Fricke dosimetry which is the radiation induced oxidization of ferrous ions (Fe^{2+}) to ferric iron (Fe^{3+}) (Fricke and Morse 1927), the color changing of Folin's phenol (Day and Stein 1950) and the radiation-induced polymerization in solutions of monomers (Hoecker and Watkins 1958). The 3D spatial information in all these methods is preserved by embedding the radiosensitive chemicals in various gel matrices which provide the necessary spatial stability of the dosimeter. Among radiosensitive gel dosimetry methods, the most widespread is that of polymer gel dosimetry, where the physicochemical principle is that of radiation induced polymerization. Free radicals produced from the radiolysis of water react with the monomers of a gel (initiation stage) creating increasingly larger macroradicals (polymerization stage) until two such macroradicals form a stable polymer macromolecule (termination stage) (De Deene 2004). The degree of polymerization, which is proportional to the absorbed dose, can then be measured as variations in the optical density of the gel (Oldham et al 2001) or more commonly as changes in the nuclear magnetic resonance relaxation characteristics (specifically transverse relaxation time T_2) of these gels (Maryanski et al 1993). An important issue in radiosensitive polymer gels is to ensure absence of any amount of oxygen, which readily scavenges free radicals produced from the radiolysis of the water, thus inhibiting the polymerization process. Thus, polymer gel dosimeters are either produced in anoxic conditions (anoxic gels) (Maryanski et al 1993), or include antioxidant agents (normoxic gels) (Fong et al 2001, Hurley et al 2005).

Radiosensitive gels have been extensively studied and successfully employed in clinical dosimetry, mainly in radiation therapy (external, internal and brachytherapy) for treatment planning verification and quality control, e.g. (Maryanski et al 1996, De Deene et al 1998, Wuu et al 2002, Gear et al 2006). In the range of energies of external beam radiotherapy, polymer gels have proven to be tissue equivalent (Fong et al 2001, Maryanski et al 1996, De Deene 2002). Additionally, a number of experimental studies indicate that the dose response of polymer gels is independent of dose rate, energy and type of radiation, for the types of

radiation usually encountered in a radiotherapeutic clinical setup (Maryanski et al 1996, Ibbot et al 1997, Farajollahi et al 1999). More recent studies, however, report some dependence of the dose response of normoxic gels with different, clinically relevant, dose rates. Specifically, significant decrease in the dose response of normoxic gels was reported for increasing dose rates between 0.5-5 Gy/min (Bayreder et al 2006). Additionally a recent feasibility study of normoxic gels as internal dosimeters in targeted radionuclide therapy supports the existence of a dependence of the gels' response from the dose rate (Gear et al 2006). While in another study for an anoxic (BANG-2) gel (Novotny et al 2001) no dependence was observed from the dose rate, a significant trend of decreasing response with increasing energies in the range of 3 – 15 MeV. Furthermore, a study of a normoxic gel reported a more pronounced under-response at high energies, where the linear energy transfer (LET) exceeds a certain threshold (4.9 keV/μm^2) (Gustavsson et al 2004). In either case, it has been suggested that the close proximity of the interaction sites increases free radical yield, subsequently increasing the number of recombination reactions between free radicals as opposed to polymerization initiation reactions (Bayreder et al 2006, Gustavsson et al 2004).

In any case, the transfer of the method from radiotherapy to interventional radiology is not a straightforward one, as both the dose rate and the energies involved in the two fields are very different. The aim of this work is to study the potential of polymer gels for dosimetry in interventional radiology. Initial investigation presented here involves the dependence of polymer gel dose response on dose rate and photon energy at energies and dose rates relevant to interventional radiology.

2. Materials and methods

2.1 Gel Manufacture

Considering the manufacturing difficulties associated with anoxic gels that prohibit their wide clinical use, a normoxic formulation was chosen for this initial investigation (De Deene 2002) and the relative concentrations of the ingredients were determined so as to maximize the gel's sensitivity to low dose ranges based on results of previous studies (De Deene 2002, Bayreder et al 2006, Hurley et al 2005). Gels were manufactured under normal atmospheric conditions using 6%(w/w) Methacrylic Acid (MAA), 4%(w/w) gelatin (300 Bloom, Aldrich) and electrophoresis high pressure liquid chromatographic (HPLC) grade distilled water, following standard procedure (De Deene 2002). The amount of 10mM tetrakis(hydroxymethyl)phosphonium chloride (THPC) was added as the antioxidant agent and 0.05mM Hydroquinone (HQ) was added to prevent spontaneous polymerization prior to irradiation. The gels were poured into test tubes which were subsequently hermetically sealed. Care was taken not to allow any air gaps to remain in the containers which would locally inhibit polymerization and disrupt the dosimetric integrity of each gel sample. Gel phantoms were generally stored in standard refrigeration (4^{o}C) to prevent gel degradation, while they were brought to room temperature prior to irradiation and MRI scanning.

2.2 Gel Irradiation

Gel irradiation was performed on an APELEM Baccara fluoroscopy unit (DMS-Apelem group, France). Dosimetric measurements were performed using an appropriately calibrated Barracuda MPD semiconductor detector (RTI, Sweden). For initial dose response evaluation, a set of gel phantom tubes were placed, along with the detector, inside the fluoroscopy unit's irradiation field leaving a 2 cm margin off the field's edges. A constant dose rate of 5 cGy/min irradiation was delivered. At predetermined time intervals, the irradiation was halted and one gel phantom tube was removed. That way, the dose response was studied for an effective dose range of 0-45 cGy, while the least possible thermal stress was imposed on the X-Ray unit. For energy and dose rate dependence, a number of gel phantom tubes were individually placed (along with the semiconductor detector) along the centre of the irradiation

field and irradiated at a constant dose of 25cGy, for varying photon energies (i.d. kVp) and dose rates. To investigate the energy dependence a number of gel phantom tubes was individually placed (along with the semiconductor detector) along the centre of the irradiation field and irradiated at a constant dose of 25cGy with a constant dose rate of 5cGy/min with varying kVp setting from 80-110 kVp in 10 kVp intervals. To investigate the dose rate dependence a number of gel phantom tubes were individually placed (along with the semiconductor detector) along the centre of the irradiation field and irradiated at a constant dose of 35cGy and constant kVp setting of 100kVp with varying dose rates between 5-8 cGy/min in 1cGy/min intervals. The dose rate was varied by changing the distance of the samples from the X-ray tube and was verified by the multipurpose semiconductor detector prior to sample irradiation.

2.3 MRI measurements and T2 calculations

All measurements were conducted on a routine clinical 1.0T scanner (Signa Horizon LX; GE Healthcare, Chalfont St. Giles, UK) using the accompanying standard head coil. Ten coronal slices were acquired using a standard 4-echo CPMG sequence, with TE: 300, 600, 900 and 1200 ms, TR: 6100 ms, slice thickness: 3mm, slice spacing: 5mm, NEX: 2, FOV 24X18 cm and 256X192 pixels. Transverse relaxation (T_2) maps were derived assuming monoexponential relaxation process and applying pixel by pixel non-linear least squares fit (in-house developed calculation software). An example of such a map is displayed in figure 1. For each gel phantom tube, the mean and standard deviation of the transverse relaxation time T_2 was measured for a fixed size region of interest, (circular area: 84 mm^2) averaging along the slices. For consistency with related literature, results are presented considering the transverse relaxivity $R2=1/T_2$ rather than the relaxation time.

Figure 1: An indicative T_2 map of the normoxic gel phantom used in this investigation.

3. Results and discussion

3.1 Dose response

Figure 2 displays the relaxivity rate of samples irradiated at a constant dose rate (8cGy/min) with a constant kVp setting (80kVp) as a function of absorbed dose, as measured by the semiconductor detector. The insert graph displays the response in absolute proportions while the main graph is adequately scaled to reveal the details of that response. This graph reveals an apparent threshold in the gel response around 25 cGy above which the gel exhibits discernible response to irradiation. A least squares linear regression of the data points above the apparent threshold, shows an adequate linear fit (r^2 = 0.9643). The numerical details of this calibration curve are also displayed on figure 2. The sensitivity value, that is, the slope of this calibration curve ($0.02\pm0.005 s^{-1}$/cGy) is in agreement with values obtained in similar studies regarding higher doses (for example cf. Bayreder et al 2006, p. 2514).

3.2. Energy dependence

Figure 3 displays the relaxivity rate of samples irradiated at a constant dose (25cGy) with a constant dose rate (5cGy/min) at varying kVp settings and consequently at varying mean X-ray energies. The insert graph displays the response in absolute proportions while the main graph is adequately scaled to reveal the details of that response. For the dose rates examined, no significant dependence of the dose response on the radiation energy could be discerned. It should be noted that energy dependence has both experimentally and theoretically been documented in gel dosimetry but for much higher energies than those encountered in all clinical setups (Gustavsson et al 2004). Additionally, Monte Carlo studies have produced non negligible dosimetric correction factors only for very low energy photons (energy of about 25keV) (Pantelis et al 2004), while the mean energy of a typical 90kVp X-ray beam with 2.5

mm Al filtering is approximately 46 keV as verified by appropriate simulation software (Novotny and Hofer 1985).

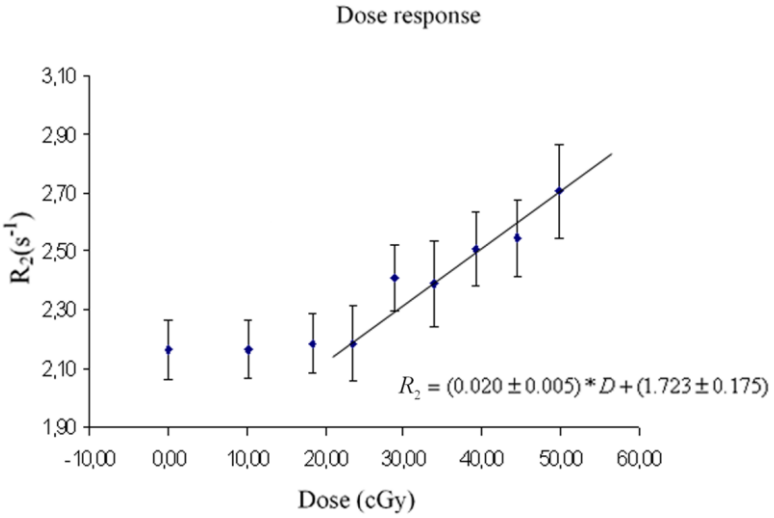

Figure 2: Dose response of the normoxic gel for photon energies of 80kVp and a dose rate of 5 cGy/min. Beyond a certain dose threshold a linear relation is observed: $R_2 = (0.020 \pm 0.005) * D + (1.723 \pm 0.175)$

Figure 3: Energy dependence of the dose response of the normoxic gel for a dose of 25 cGy delivered at a dose rate of 5 cGy/min. The dependence is negligible within the experimental error.

3.3. Dose rate dependence

Figure 4 displays the relaxivity rate of samples irradiated at a constant dose (35cGy) with a constant kVp setting (100kVp) at varying dose rates. The insert graph displays the response in absolute proportions while the main graph is adequately scaled to reveal the details of that response. The graph shows a dependence of the dose response on the dose rate, which can be modelled mathematically via a linear least squares fit (r^2=0.9774). The details of the linear model are also displayed in figure 4. From these data there appears to be an increase in the gel's response with increasing dose rates. That would suggest that reducing the dose rate would lead to decreased sensitivity and subsequent increase in the dose threshold beyond which discernible dose response of the gels is observed.

Figure 4: Dose rate dependence of the dose response of the normoxic gel, for photon energies of 100kVp and a total dose of 35cGy. A linear relation is observed: $R_2 = (0.055 \pm 0.026) * \dot{D} + (1.495 \pm 0.168)$

Although the dependence of the dose response is rather weak (as demonstrated from the slope of the linear fit), so that it could be considered practically negligible, there is a more fundamental issue to be explored in these results. Specifically, the observed dependence seems to contradict existing evidence of dose rate dependence in dosimetric gels. There are a number of studies (Bayreder et al 2006, Gear et al 2006, De Deene 2004) which report a decrease in gels' response with the increase of the dose rate. In gel dosimetric studies

conducted at dose rates orders of magnitude higher than those encountered in interventional radiology and reproduced in this study, it has been argued that an increase in dose rate leads to a closer proximity of the interaction sites, increasing free radical yield and thus increasing the number of recombination reactions between free radicals as opposed to polymerization initiation reactions (De Deene 2004). In fact the chemical processes that initiate and terminate the polymerization have been quite adequately explored (De Deene 2002). The process begins with the radiolysis of water molecules in the gel into 2 radicals $R^{\bullet}$. The radicals initiate polymerization reactions with the monomers M_n at the site of their formation:

$$R^{\bullet} + M_n \rightarrow 2RM_n^{\bullet} \quad (1)$$

and

$$RM_n^{\bullet} + M_m \rightarrow 2RM_{n+m}^{\bullet} \quad (2)$$

This process can end with a number of termination reactions, which all lead to stable polymers:

$$R^{\bullet} + R^{\bullet} \rightarrow 2R = H_2O \quad (3)$$

or

$$R^{\bullet} + RM_n^{\bullet} \rightarrow H_2O + M_n \quad (4)$$

or

$$RM_n^{\bullet} + RM_m^{\bullet} \rightarrow H_2O + M_{n+m} \quad (5)$$

or

$$RM_n^{\bullet} + RM_m^{\bullet} \rightarrow H_2O + M_n + M_m \quad (6)$$

While antioxidants are included in the gel, it is impossible to achieve 100% oxygen scavenging. Thus, these termination processes are antagonistic to oxygen scavenging of free radicals, as well as polymerization inhibition reactions by hydroquinone (De Deene 2002), which is added to prevent spontaneous polymerization in the absence of radiation (Fong et al. 2001). To overcome these competing mechanisms a minimum dose and dose rate is required. Previous studies have been conducted at dose rates in the range of several Gy/min (Bayreder et al 2006, Gear et al 2006, De Deene 2004), while this paper focuses on interventional radiology applications, typically at dose rates of a few cGy/min (Wagner et al 1994, Meyer et al 2001, Vano et al 2001). At these relatively very low dose rates it is possible that these competitive mechanisms produce an observable effect in gel response. When the dose rate becomes significantly higher, these mechanisms become saturated and the previously described effect of increased recombination reactions due to closer proximity of the interaction sites becomes dominant. While it is beyond the scope of this preliminary study, quantitative investigation of these processes presents an interesting challenge for further research.

4. Conclusions

The aim of this paper is to investigate the feasibility of normoxic gel dosimetry in interventional radiology. Preliminary results indicate that the specific normoxic gel studied exhibits a linear dose response for doses in the range of 25-55 cGy, although there appears to be a lower dose threshold of around 25 cGy below which the gel does not respond to irradiation. The dependence of the dose response to photon energies of X-rays produced from

voltage between 80-110 kVp is negligible (within the experimental error). However, experimental data indicates a linear dependence of dose response to dose rate in the range of 5-8 cGy/min. While at high dose rates the dosimetric response of the gels increases as the dose rate decreases (Bayreder et al 2006, Gear et al 2006, De Deene 2004), oxygen scavenging of free radicals and polymerization inhibition reactions by hydroquinone seem to reverse this trend in very low dose rates. This may create potential source of error in dosimetry of complex interventional radiology procedures where multiple irradiation fields are employed with an overall irradiation of variable dose rate. Further work is therefore required to quantify the antagonistic mechanisms which lead to this reversal of dose response at low dose rates and investigate the possibility of correction - calibration procedures, before normoxic gel dosimetry can be routinely applied for 3D dosimetry in complex interventional radiology procedures.

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