A model for post-irradiation effects in polymer gel dosimeters

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1. Introduction

Soon after the introduction of polymer gel dosimeters in 1993 [1], edge enhancing effects were noticed in these gels near high dose gradient regions at higher dose levels [2] (typically doses beyond the linear dose-R2 relationship). Our research group has also reported on this phenomenon in several studies [3–5]. It is believed that due to a depletion of monomers in the high dose region after irradiation, a net diffusion occurs of unreacted monomer into this region where it reacts with long-living macroradicals. As a result, dose ‘overshoots’ can be observed in the gel-measured dose distribution. However, this hypothesis has never been substantiated.

When examining the dose-R2 relation post-irradiation, two types of instabilities may be observed [6,7]: (a) a polymerization reaction and (b) the gelation process of gelatin. The polymerization reaction post-irradiation is also believed to be due to the creation of macroradical products.

A mathematical model is proposed that links the temporal instability to the overshoots near high dose gradients. This model is supported by experimental data. The work is still in progress.

2. Materials and methods

The input parameters for the model are: (i) the monomer diffusion coefficient(s) in gelatin (for a certain gelatin concentration), (ii) a 1D absolute dose distribution, (iii) the dose-R2 relationship (mono-exponential) and (iv) its instability as a consequence of post-irradiation polymerization. All input parameters are experimentally determined (see further). The model predicts the initial distribution of monomer, polymer and macroradicals, and simulates the evolution of the measured R2 (and dose) distribution in time.

Most parameters needed as input for the simulations were derived from a separate stability study. For this, four batches of polymer gel were produced (according to the procedure described in [3]), with different gelatin concentrations (0 – 3 – 6 – 12 %). Each batch was filled in a number of test tubes, and each test tube was irradiated with a different known dose (from 0 to 50 Gy). Irradiation was done with a 10x10cm² field of 6MV photons (SLi18, Elekta, Crawley, UK), with source-to-surface distance = 95 cm and depth = 5 cm. During irradiation, the laterolateral field penumbra was located at the center of the test tube. After irradiation, all test tubes were scanned (Symphony 1.5T, Siemens, Erlangen, Germany) lengthwise at regular time intervals for 4 weeks.

The monomer diffusion coefficient was determined by diffusion measurements with a benchtop NMR system (Minispec mq20, Bruker, Rheinstetten, Germany). For these measurements, all samples were prepared with deuterium oxide instead of water to avoid contribution of diffusing water protons.
3. Results

The dose sensitivity (slope of the dose-R2 curve) as a function of time for each gel batch is displayed in figure 1. A relation is observed between gelatin concentration and gel sensitivity. An exponential function of the form $Sl = Sl_0 + \Delta Sl_{inf} \cdot (1 - \exp(-\text{time}/T_p))$ is fitted to the data to describe the evolution of the sensitivity with post-irradiation time. From this function, the slope at time = 0 ($Sl_0$), the total increase in slope ($\Delta Sl_{inf}$) and the macroradical time constant ($T_p$) are extracted. Figure 2 shows the macroradical time constant as a function of gelatin concentration. The values for gelatin concentrations of 5% and 10% are adapted from [8]. Measured diffusion coefficients are given in table 1.

![Figure 1](image1.png)  
**Figure 1.** Dose sensitivity as a function of time for each gel batch.

![Figure 2](image2.png)  
**Figure 2.** Macroradical time constant as a function of gelatin concentration.

| Gelatin Concentration | Acrylamide | Acrylamide + Bisacrylamide |
|-----------------------|------------|---------------------------|
| 3% gelatin            | (927 ± 41).10^{-12} m²/s | (543 ± 13).10^{-12} m²/s |
| 6% gelatin            | (610 ± 46).10^{-12} m²/s | (478 ± 16).10^{-12} m²/s |

Table 1. Diffusion coefficients of monomer(s) for different gelatin concentrations.

Figure 3 shows R2 profiles measured at different times post-irradiation in the 3% gelatin gel, irradiated with 15 Gy at the isocenter. The over-response of the gel near the penumbra becomes apparent after a few hours. The increase in R2 in the high dose region is a consequence of the post-irradiation polymerization. Figure 4 shows a simulation of the case shown in figure 3. For the simulation, diamond detector measurements of the 6MV 10x10 cm² field were used as input for the 1D dose distribution. The measurements also showed a lower amplitude of the dose overshoot for higher concentrations of gelatin. Another experiment (result not shown) demonstrated that the dose overshoot amplitude is independent of the dose rate (between 50 and 500 cGy/min) during irradiation.
4. Discussion

The stability study demonstrated a decrease in the dose sensitivity for increasing concentrations of gelatin. Diffusion measurements showed an decrease of the monomer diffusion constant for increasing gelatin concentrations. It is likely that, due to the restricted diffusion of monomer in gels with a higher gelatin concentration, these monomers have more difficulty to reach the reactive sites on the polymer radicals, thus resulting in a decreased dose sensitivity. A correlation was also found between the gelatin concentration and the macroradical time constant, indicating the involvement of gelatin in the polymerization process.

When comparing measured R2 profiles with simulations, a strong correlation is found. This confirms the hypothesis that an inward diffusion of monomers into the high dose region is responsible for the gel's overresponse when irradiated to high doses. With the theoretical model, a connection is demonstrated between the temporal instabilities (post-irradiation polymerization) and the spatial instabilities (overshoots) in polymer gels.

Diffusion measurements indicated a higher diffusion coefficient for acrylamide, in comparison to the combination acrylamide + bisacrylamide. This indicates a low diffusion coefficient for bisacrylamide. Also, bisacrylamide is depleted at a higher rate than acrylamide [9,10]. For these reasons, it can be expected that bisacrylamide plays only a minor role concerning the overshoots and therefore the diffusion coefficient of acrylamide was used in the simulations.

Some features could further improve the theoretical model. Results indicate that it is likely that the monomer diffusion coefficient is influenced by the local degree of polymerization. This will be investigated. Also, the model assumes a mono-exponential dose-R2 relationship (instead of bi-exponential) and a linear relation between measured R2 and polymer concentration. The change in R2 as a consequence of gelation of gelatin is also not incorporated in the model.
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