Experimental investigations of polymer gel dosimeters

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Abstract
Polymer gel materials continue to show promise as 3D dose verification tools in radiotherapy, particularly given the ever-increasing complexity of modern radiation therapy treatment techniques. To date different polymer gel formulations have been explored as potential 3D dosimeters, each with different characteristics. Although the fundamental processes (e.g. polymerization) occurring in many of these dosimeters share common principles, experimental and environmental factors (e.g. gel irradiation temperature) can differentially affect the different polymer gel dosimeters. This dependence can further be amplified or suppressed depending on the imaging modality used to extract the dose information from the polymer gel. This paper reviews the experimental investigations of the fundamental processes occurring in irradiated polymer gel dosimeters, including monomer consumption, polymerization, gelation, and gel ageing. The experimental and environmental factors which affect these processes during, and post, gel irradiation are also discussed. Finally, since the body of literature pertaining to basic investigations in polymer gel dosimetry is ever expanding, some effort is made to catalogue both published experimental work as well as unexplored research areas in the field.

1. Introduction
During the past 10 years there has been considerable and continually increasing interest in the use of polymer gel materials for 3D dose verification in radiation therapy [1-3]. The basic fundamental principle of polymer gel dosimeters involves the radiation-induced polymerization of monomer (and often co-monomer) species suspended in a gelatin matrix. Radiation induced polymerization creates long-chained polymers that are spatially retained in the gelatin matrix, thus allowing for the extraction of 3D dose information from the polymer gel dosimeters. To date, a number of different polymer gel compositions have been investigated (see table 1 below) as potential materials for 3D dose verification. Furthermore, the requirement for highly accurate dosimetry has necessitated and understanding of the effect of experimental and environmental parameters on the accuracy, precision, and robustness of polymer gel dosimetry. The experimental and environmental parameters may have differential effects on the different polymer gel compositions and these effects may be more or less pronounced in the overall system depending on the imaging modality used for dose information extraction. Hence, it can be argued that while the current state of knowledge in polymer gel dosimetry is expanding, so is the complexity of polymer gel dosimetry, particularly to the non-polymer-gel-specialist. Hence this review aims to help assess the current status of our knowledge of the fundamental processes occurring in polymer gel dosimeters. As any review must necessarily be, this is
a partial summary of the extensive experimental evidence concerning both the fundamental processes occurring in polymer gel dosimeters and the experimental and environmental parameters which affect these processes. This review also serves to catalogue the current state of completed experiments on basic polymer gel investigations.

2. Polymerization and monomer consumption in polymer gel dosimeters

Central to the functioning of any polymer gel system is the radiation-induced polymerization of the monomer species present in the gel. Traditionally (e.g. in polyacrylamide gels – PAG) radiation induces polymerization of a monomer (acrylamide) and a crosslinker (bis-acrylamide, bis). Several more recent polymer gel dosimeters utilize a single monomer species (e.g. methacrylic acid). In either monomer / co-monomer or single monomer systems, a number of factors affect the rate of polymerization, or rate of monomer consumption. These factors are described below.

2.1. Polymer gel composition

2.1.1 Gel sensitivity. The effect of polymer gel composition on net gel sensitivity to radiation has been studied by a large number of researchers [4-20]. Without doubt the choice of monomer (and co-monomer) within the gel plays a large role in the net sensitivity of the polymer gel to radiation. Early on, Maryanski et al [18] utilized MRI to establish that in traditional PAG dosimeters, varying the relative fractions of acrylamide and bis-acrylamide has a dramatic effects on gel sensitivity to radiation. 

![Figure 1](image_url)

**Figure 1.** (a) Affects of the relative mass fraction of bis-acrylamide to total monomer (%C) on dose sensitivity [18] (b) Average number of acrylamide to bis-acrylamide molecules available for reaction as a function of %C and dose - illustrates differential consumption rates of acrylamide and bis [13], (c) Rate of acrylamide and bis consumption as a function of total monomer concentration - $D_{1/2}$ relates to the decay constant in the exponential fit to the dose response curve [15].
radiation (figure 1a). More recently, FT-Raman spectroscopy has been used in the direct monitoring of monomer consumption and polymer formation in polymer gel dosimeters [19,20,13]. Baldock et al were the first to illustrate the use of FT-Raman spectroscopy for the study of monomer consumption in irradiated polymer gel dosimeters. Baldock et al also showed the differential rate of consumption of acrylamide relative to bis-acrylamide in irradiated PAG dosimeters. Jirasek et al [13] used Raman spectroscopy to extend the work of Baldock and directly monitor the radiation-induced consumption of acrylamide and bis-acrylamide for varying gel compositions (figure 1b). It was found that bis is consumed at a greater rate than acrylamide for all gel compositions studied, resulting in a heterogenous polymer structure [19]. Fuxman et al (10) subsequently provided mathematical models for acrylamide and bis-acrylamide consumption. McAuley et al (in [3]) have further pointed out that, in fact, bis-acrylamide acts as a poor crosslinker, readily reacting through individual molecule primary cyclizations to create 7 – membered bis-acrylamide rings. Hence, longer molecule crosslinkers may offer improved-performance dosimeters. Lepage et al used Raman spectroscopy and modeling to show that increasing the total monomer / crosslinker fraction in PAG gels leads to a lower rate of monomer / crosslinker consumption (figure 1c) [15]. This effect has been attributed to a possible increase in termination (through cyclization) of bis-acrylamide with increasing initial monomer concentration [21,22]. Changing the relative fraction of monomer / crosslinker has further implications for the polymer “particle” size created in irradiated dosimeters, as discussed by Maryanski et al [23], and polymer density, as discussed by Hilts et al [5]. These two facts have direct consequences for gel sensitivity when imaged with optical computed tomography (OCT) and x-ray computed tomography (XCT).

Although PAG remains the most studied dosimeter in these regards, other dosimeters have been investigated (see table 1).

Notably, Fong et al [24] presented the first preliminary studies of a normoxic (i.e. dosimeter manufactured under normal atmospheric conditions and containing an oxygen scavenger) methacrylic polymer gel dosimeter (termed MAGIC). This dosimeter contains a single monomer species, ascorbic acid as oxygen scavenger, CuSO₄ as an initiator of O₂ scavenging, and hydroquinone as free radical scavenger. De Deene et al [11] presented a thorough study on the effects of the above gel constituents on ΔR2, the change in MRI R2 relaxation rate with a given (5 Gy) dose (figure 2). Furthermore, De Deene et al [11] showed preliminary results for simplified dosimeters (MAGAS, i.e. MAGIC without CuSO₄ and hydroquinone, and PAGAS, an acrylamide, bis, and ascorbic acid

![Figure 2](image.png)

**Figure 2.** Change in R2 for MAGIC gels composed of varying amounts of copper(II) sulphate and hydroquinone and irradiated with a dose of 5 Gy. (a) ascorbic acid concentration = 1 mM. (b) ascorbic acid concentration = 5 mM (11).
Table 1. Compositions of the most widely studied polymer gel dosimeters. N = normoxic, A = anoxic, G = gelatin, E = either (gelatin or agarose based).

|                   | Anoxic / Normoxic | Gelatin / Agarose | Acrylamide | N,N’-methylene bis acrylamide | Methacrylic acid | Acrylic acid | N-methacryloyl | N-vinylpyrrolidone | 2-hydroxyethyl methacrylate | Ascorbic acid | Hydroquinone | Copper(II) sulphate | THPC\(^1\) | THPS\(^2\) |
|-------------------|-------------------|-------------------|------------|-------------------------------|------------------|-------------|--------------|-----------------|-----------------------------|----------------|--------------|---------------------|----------|----------|
| PAG               | A                 | E                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| nPAG\(^3\)       | N                 | E                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| PAGAT\(^4\)      | N                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| PAGAS             | N                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| MAGIC             | N                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| nMAG              | N                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| MAGAS             | N                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| MCA               | A                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| MAGAT             | N                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| Methacrylate      | A                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| VIPAR             | A                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| HEA               | A                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| HEMA              | A                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| ACA               | A                 | G                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |
| PRESAGE           | N                 | •                 | •          | •                             | •                | •           | •            | •               | •                           | •              | •            | •                   | •        | •        |

1. tetrakis hydroxymethyl(phosphonium) chloride  
2. bis [tetrakis hydroxymethyl(phosphonium) sulphate]  
3. nPAG has been manufactured with either THPC or THPS  
4. Some inconsistencies exist in the literature pertaining to the naming of PAGAT and nPAG.

dosimeter), as well as dosimeters based on a range of antioxidants. Of the antioxidants tested, tetrakis hydroxy(methyl) phosphonium chloride (THPC) showed the greatest rate of oxygen scavenging and the most promise as an oxygen scavenger for polymer gel dosimeters. The oxygen scavenging ability of THPC is based upon the conversion of THPC to THP (tetrakis (hydroxymethyl) phosphonium and the subsequent reaction with molecular oxygen, as described in equations 1 and 2\[^{25}\].

\[
\begin{align*}
\text{(HOCH}_2\text{)}_4\text{POH} & \leftrightarrow \text{(HOCH}_2\text{)}_3\text{P} + \text{HCHO} + \text{H}_2\text{O} \\
\text{(HOCH}_2\text{)}_3\text{P} + 0.5\text{O}_2 & \rightarrow \text{(HOCH}_2\text{)}_3\text{P} = \text{O} 
\end{align*}
\]

Since the initial investigation of De Deene et al\[^{11}\], a number of polymer gel dosimeter formulations have been investigated utilizing, mainly, MRI and XCT (see references in caption of table 2). Most investigations have at least partly centred on the optimization of the formulations for maximized polymer gel sensitivity to radiation. Table 2 provides a comparison of the optimized dose sensitivities of different formulations of polymer gel dosimeters to radiation, as determined by MRI, OCT, or XCT. It is noted that these types of comparisons are difficult to compile, due in part to the different methods used to determine gel sensitivity. However, the table is included in order to give some sense of what, to date, has been studied and the experiments remaining to be conduced.
Table 2. Examples the of the sensitivity to radiation of different polymer gel formulations. Taken from references [1,3,4,6,7,9,11,14,16,23,25-29]. g = gelatin based gel, a = agarose based gel.

| PAG | nPAG | PAGAT | PAGAS | MAGIC | nMAG | MAGAS | MCA | MAGAT | Methacrylate | VIPAR | HEA | HEMA | ACA | PRESAGE |
|-----|------|-------|-------|-------|------|------|-----|-------|-----------|-------|-----|------|-----|---------|
| g   | a    | g     | a     | g     | g    | g    | g   | g     | a         | a     | a   | a    | a   | --      |
| MRI (s$^{-1}$Gy$^{-1}$) |   |   |   |   |   |   |   |   |   |   |   |   |   |   |
| 0.33 | 0.475 | 0.12 | 0.192 | 0.005 | 0.008 | 0.732 | 2.1 | 0.35 | 1.19±0.05 | 3.2 | 0.21 | 0.05 | 0.002 | 0.35±0.06 |

| XCT (H/Gy) | OCT (O/DGy) |   |   |   |   |   |   |   |   |   |   |   |   |   |
| 0.86±0.04 | 0.38±0.07 | 0.86±0.8 | 0.95 | 0.498±0.003 | 0.46±0.002 | 0.013 |

2.1.2 Attenuation properties. An often cited advantage of polymer gel dosimeters over other types of dosimetry systems is their inherent tissue equivalence. However, as new polymer gel dosimeter formulations are investigated, the tissue equivalence, or attenuation properties, of these dosimeters must be verified, particularly for formulations using larger quantities of antioxidants containing moderately high Z elements (e.g. THPC). The attenuation properties of different polymer gel dosimeters has been studied by several workers, e.g. [40,41] and are often quoted within a given irradiation energy range. Table 3 summarizes the current known attenuation properties of polymer gel dosimeters.

2.2. Irradiation energy – LET
Polymer gel dosimeters are attractive systems for applications in particle therapy (i.e. high incident particle linear energy transfer, LET) due to their inherent high spatial resolution and the high dose gradients associated with these irradiation modalities. Several workers have investigated the response of polymer gel to incident particle LET [4,25,43-46]. In general, incident particle LET will alter the response of polymer gel dosimeters, virtually irrespective of the formulation of dosimeter used. This effect can be explained by the dose deposition characteristics of high LET (i.e. proton and above) radiations. As shown by Jirasek and Duzenli [45], calculations can be made for the deposition of dose, due to delta rays ejected from the primary incident particle track, as a function of distance from that track (figure 3). It was shown that the high delta ray dose deposited close to the track of the incident particle saturates the dosimeter, leading to an under-response of polymer gel to high LET radiation dose. Experimental verifications of this under-response have been cited in several works
Due to the high delta ray doses close to the track of the incident particle, future formulations of polymer gel can also be expected to exhibit similar behaviour.

Table 3. Attenuation properties of polymer gel dosimeters. Shown is the % difference between the mass attenuation coefficient of the given polymer gel and water. Taken from references [40-42].

|                | PAG  | nPAG | PAGAT | PAGAS | MAGIC | nMAG | MAGAS | MCA  | MAGAT | Methacrylate | VIPAR | HEA | HEMA | ACA | PRESAGE |
|----------------|------|------|-------|-------|-------|------|-------|------|-------|--------------|-------|-----|------|-----|---------|
| <50 keV        | g    | g    | g     | g     | g     | g    | g     | g    | g     | <0.97        | 1.02  |     |      |     |         |
| 0.1-1 MeV      | g    | g    | g     | g     | g     | g    | g     | g    | g     | >0.99        | 1.03  |     |      |     |         |

2.3. Irradiation dose rate
Several investigators have studied the effects of irradiation dose rate on various formulations of polymer gel dosimeters [4,46]. As noted in De Deene et al [4], radiation-induced polymerization proceeds in several steps. Initiation of polymerization proceeds through the reaction of monomer with radiation induced water radicals. It is the production of the water radicals which is dependent upon the irradiation dose rate. Polymer propagation proceeds via the formation of a long chain polymer. The

**Figure 3.** Delta-ray dose deposited in a polymer gel as a function of distance from the incident proton particle track. Illustrated is the saturation effect (i.e. high saturation doses) close to the particle track, leading to dosimeter under-response (relative to photon irradiated polymer gels).
termination of polymerization occurs via, e.g., the combination of two polymer radicals. The equilibrium concentration of water radicals depends on the rate of radical formation (via incident radiation dose rate) and the loss of water radicals via polymer propagation or termination. The rate of propagation and termination is dependent on the radical concentration, while the creation of water radicals is generally independent of radical concentration. Hence, these two factors combine to form a dose rate dependent polymer gel dosimeter. The extent of polymer gel dose rate dependence depends primarily on the reactivity of the polymer gel constituents, and also the gelatin concentration. Table 4 summarizes the dependence of various polymer gel dosimeters on dose rate.

Table 4. Largest change in slope, expressed as a % difference, in polymer gel dose response when irradiated with incident photon dose rates between 10 and 400 cGy/min. Note: results for PAG, nPAG, and ACA are within errors bars of experiment. g = gelatin based gel. Taken from [4,46].

|          | PAG | nPAG | PAGAT | PAGAS | MAGIC | nMAG | MAGAS | MCA | MAGAT | Methacrylate | VIPAR | HEA | HEMA | ACA | PRESAGE |
|----------|-----|------|-------|-------|-------|------|-------|-----|-------|--------------|-------|-----|------|-----|---------|
| % diff   | 3%  | 5%   | 16%   |       |       |      |       |     |       |              |       |     |      |     |         |

2.4. Polymer gel irradiation temperature
The temperature of polymer gel at the time of irradiation can have an effect on the resulting dose response of the system due to the general influence of temperature on reaction kinetics. Temperature influences not only the reaction rates of the polymerization reactions, but also the viscosity of the polymer gel. The gel temperature at irradiation can thus have an influence on the dose response of the polymer gel [3,4,47]. Furthermore, the extent of effects of temperature on polymer gel dose response can be influenced by the composition of the polymer gel (e.g. THPC – based PAG gels exhibit an overall greater viscosity than do traditional PAG dosimeters) [25]. Hence, it is expected that the different polymer gel formulations will be effected by irradiation temperature to different extents. For example, De Deene et al [4] (using MRI) and Hilts et al [45] (using XCT) have observed no significant alterations for PAG dose response for gels irradiated between 4 - 20 °C. Similar results were obtained using MRI for nPAG dosimeters. However, nMAG R2 dose response (measured with MRI) can be influenced by temperature to the order of 0.034 s⁻¹ Gy⁻¹ K⁻¹. It has been pointed out that effects of gel temperature during irradiation should be taken into consideration during a given experiment since the polymerization reaction is exothermic and hence a non-homogenous temperature distribution can be obtained within a polymer gel during irradiation [46].

3. Gel ageing , Gelation
Baldock et al have shown that polymerization of PAG dosimeters manufactured under anoxic conditions can proceed for up to 12 hrs post polymer gel irradiation [in 1]. In addition to this process, the gelatin matrix can “age” over time, thus altering the characteristics of the polymer gel dosimeter. Both the imaging time post-irradiation, and the gel temperature at imaging can affect the overall results of any given experiment. These effects will also be more or less pronounced if different imaging modalities are used to extract the dose information from the polymer gel. Two parameters affected by gel aging / gelation are discussed below.
Figure 4. Changes in (a) slope and (b) baseline R2 values as a function of imaging time post irradiation. Shown are a range of polymer gel formulations. Taken from reference [49].

3.1. Post irradiation time
The amount of time between gel irradiation and gel imaging can affect gel imaging results and has been studied by a number of workers [3,4,6,9,25,31,47,49,50]. De Deene et al [31] identified two types of post-irradiation instability. The first instability relates to a continuation of the polymerization process (post-irradiation) due to the presence of long lived radicals which propagate the polymerization process. Direct monitoring of this post-irradiation instability has been observed by Baldock et al [1] using FT-Raman spectroscopy. In their study, Baldock et al monitored the post-irradiation consumption of monomers in PAG gels and noted that monomers continue to be consumed for up to 12 hrs post polymer gel irradiation. De Deene et al showed that this instability translated to a change in the slope of the MR R2 vs dose curve of these gels and that the changing slope stabilizes after 12 hrs post gel irradiation [31]. Thus, if polymer gels are imaged a minimum of 12 hrs post irradiation, systematic error due to this effect should be minimized.

De Deene et al also identified a second long term instability in irradiated polymer gels, relating to the long-term gelation process of the gelatin in the system. Generally speaking, they found that the long term (e.g. 30 day) aging process of gelatin once renatured below 35 °C affects the intercept of the R2 vs dose response curve (i.e. R2o), while leaving the slope of this curve unaltered. Gelatin is a network of biopolymer chains composed of links of collagen (∼280 nm in length). Each link of collagen is composed of three polypeptide chains, wound into a left-handed helix. The chain is wound into a right-handed helix structure [51]. A number of studies have shown that gelation is extremely rapid initially (first few minutes of solution being below 35 °C) but evolves much slower after this phase, never truly reaching equilibrium [52,53]. Furthermore, monomer constituents (e.g. acrylamide or methacrylic acid) can influence the gelatin structures, thus altering the measured R2o values. Representative changes in R2 and R2o are shown in figure 4.

The effects of gelation and gel aging are not observed (within experimental uncertainty) when polymer gels are imaged with XCT. Hilts et al [47] showed that the slope of the XCT number (ΔN<sub>CT</sub>) vs dose response curve for PAG gels remained constant when the polymer gel was imaged over several days. Similar results were obtained by Jirasek et al [25] for nPAG gels imaged up to 5 days post-irradiation. In both studies, a background image was subtracted from the original gel image and results were normalized to 0 Gy. Hence, the information of the intercept of the ΔN<sub>CT</sub> vs dose response curve was not recorded in these studies.

Krstajic et al [in 3] have shown dose response reproducibility curves for PRESAGE gels imaged with OCT. Their preliminary results indicate some variation in the slope of the optical density (OD) vs dose response curve for PRESAGE gels imaged up to 9 weeks post irradiation. No explanation was given for this effect. Table 5 summarizes the effects of post-irradiation time on polymer gel stability.
3.2. Polymer gel imaging temperature
The mobility of monomers, polymer structures, and water in the gelatin matrix of a polymer gel are affected by the temperature of the polymer gel since the structure of the gelatin matrix (e.g., its viscosity) depend to some degree on temperature. It has been shown that the dose sensitivity of PAG and nMAG polymer gel dosimeters (as measured using MRI) varies as a function of temperature, leading to a possible 7 – 8% / °C dose inaccuracy for PAG and a 4 – 5% / °C dose inaccuracy for nMAG dosimeters [in 3]. Further MRI imaging temperature studies have yielded similar results [54]. XCT studies of polymer gel imaging temperature during CT scanning have yielded a slight variation in the slope of the \( \Delta N_{CT} \) vs dose response curve [47].

**Table 5.** Change in slope (in %) of the polymer gel dose response curve as a function of imaging time post irradiation. Taken from [3,4,6,7,11,25,31,47,50].

|          | PAG | nPAG | PAGAT | PAGAS | Magic | nMAG | MAGAS | MCA | MAGAT | Methacrylate | VIPAR | HEA | HEMA | ACA | PRESAGE |
|----------|-----|------|-------|-------|-------|------|-------|-----|-------|--------------|-------|-----|------|-----|--------|
| MRI      | 43  | 35   | 5     | -20   | -21   | 8.5  | 9.5   | 17  |       |              |       |     |      |     |        |
| OCT      |    |      |       |       |       |      |       |     |       |              |       |     |      |     |        |

4. Conclusion
A review has been given of the experiments highlighting the fundamental processes occurring in irradiated polymer gel dosimeters, as well as the environmental factors which affect these processes. The current status of experimental work in this field has been catalogued. Over the past decade of polymer gel dosimetry research, experimental methods have been established and are now available for the study of a large number of polymer gel dosimeters. These established experimental methods will help to ensure that any new polymer gel dosimeters developed for radiation therapy dose verification will meet clinical requirements.

5. References
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