Historical overview of the development of gel dosimetry: a personal perspective

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1. Introduction
Over many years individuals have endeavoured to measure absorbed radiation dose distributions using gels. As long ago as 1950, the radiation-induced colour change in dyes was used to investigate radiation doses in gels [1]. Further, in 1957 depth doses of photons and electrons in agar gels were investigated using spectrophotometry [2]. Gel dosimetry today however, is founded mainly on the work of Gore et al in 1984 [3] demonstrated that changes due to ionising radiation in Fricke dosimetry solutions [4], developed in the 1920’s, could be measured using nuclear magnetic resonance (NMR).

2. Fricke Gel Dosimeters
Gore et al investigated [3] the nuclear magnetic resonance (NMR) relaxation properties of irradiated Fricke or ferrous sulphate dosimetry solutions [4] showing that radiation-induced changes, in which ferrous (Fe2+) ions are converted to ferric (Fe3+) ions, could be quantified using NMR relaxation measurements. In 1986 Appleby et al [5] reported that Fricke dosimetry solutions dispersed throughout a gel matrix could be used to obtain three-dimensional (3D) spatial dose information using magnetic resonance imaging (MRI). It was subsequently shown that irradiated Fricke-type gel dosimeters did not retain a spatially stable dose distribution due to ion diffusion within the irradiated dosimeters [6]. Fricke solutions with various gelling agents such as gelatine, agarose, sephadex and polyvinyl alcohol (PVA) were investigated along with chelating agents such as xylenol orange (XO) to reduce diffusion. Numerous authors subsequently published results of their work to inhibit the ion-diffusion with limited success and summarised by Baldock in 2001 (Table 1) [7]. By the early 1990’s the diffusion problem was considered to be a significant one in the advancement of gel dosimetry.

3. Polymer Gel Dosimeters
Polymer systems for the use of radiation dosimetry were first proposed as early as 1954, where Alexander et al [8] discussed the effects of ionising radiation on polymethylmethacrylate. Following this, Hoecker et al [9] in 1958 investigated the dosimetry of radiation-induced polymerisation in liquids, and in 1961 Boni [10] used polyacrylamide as a gamma dosimeter. Much later in 1991, Audet et al [11] reported changes in NMR transverse relaxation measurements of irradiated polyethelene
oxide. In 1992, Kennan et al [12] reported on NMR longitudinal relaxation studies performed on an irradiated aqueous solution of N,N'-methylene-bis-acrylamide and agarose, which showed that the relaxation rates increased with absorbed dose.

In 1992 a new gel dosimetry formulation was proposed, which was based on the polymerisation of acrylamide and N,N'-methylene-bis-acrylamide (bis) monomers infused in an aqueous agarose matrix [13]. This system was given the acronym BANANA due to the use of the chemical components (bis, acrylamide, nitrous oxide and agarose) [14]. This type of gel dosimeter did not have the associated diffusion problem of Fricke gels and was shown to have a relatively stable post-irradiation dose distribution. The polymerisation reaction occurred by cross-linking of the monomers induced by the free radical products of water radiolysis. In 1994 the BANANA formulation was refined [15] by replacing agarose with gelatine and given the acronym BANG (bis, acrylamide, nitrogen and aqueous gelatine), the first of a series of new polymer gel formulations. In 1994 this formulation was patented [16] and became commercially available through MGS Research Inc. as BANG®. Subsequently, due to the naming of the commercial product, PAG [17] became the polymer gel dosimeter acronym of choice for most authors.

Numerous authors subsequently published results of work investigating different compositions and formulations of polymer gel dosimeters some of which were summarised by Lepage et al (Table 2) [18].

Although polymer-type dosimeters did not have the diffusion limitations of Fricke-type gel dosimeters, there was another significant limitation in their use. Due to the nature of their free radical chemistry, polymer gel dosimeters were susceptible to atmospheric oxygen inhibition of the polymerisation processes. As a result, these gel dosimeters had to be manufactured in an oxygen-free environment [17], such as in a glove box pumped with nitrogen gas. Along with the use of potentially toxic chemicals [19], this was a significant limitation in the introduction of gel dosimetry into the clinic.

4. Normoxic Polymer Gel Dosimeters

A significant development in the field of gel dosimetry occurred when results of using an alternative polymer gel dosimeter formulation were published by Fong et al in 2001 [20]. This new type of polymer gel dosimeter, know as MAGIC gel, bound atmospheric oxygen in a metallo-organic complex thus removing the problem of oxygen inhibition and enabling polymer gels to be manufactured on the bench-top in the laboratory. This created what was to be known as a normoxic gel dosimeter, compared with the previous PAG formulation which subsequently became known as a hypoxic gel dosimeter.

The MAGIC polymer gel formulation consisted of methacrylic acid, ascorbic acid, gelatine and copper. The principal behind the MAGIC gel is in the ascorbic acid oxygen scavenger. Ascorbic acid binds free oxygen contained within the aqueous gelatine matrix into metallo-organic complexes and this process is initiated by copper sulphate. It was subsequently shown by De Deene et al in 2002 that other antioxidants could be used in the manufacture of normoxic gels [21] including tetrakis (hydroxymethyl) phosphonium chloride, having first been suggested to Baldock by Billingham in 1996 [22].

Numerous authors subsequently published results of work investigating different compositions and formulations of normoxic polymer gel dosimeters and were recently summarised by Senden (Table 3) [23].

Recent work has included the development of less toxic polymer gels [23].
Table 1. Summary of Fricke gel dosimeter diffusion measurements in the literature in Baldock et al [6].

|   | Diffusion Coefficient ($10^{-3} \text{ cm}^2\text{h}^{-1}$) | Gel Type & Concentration (%) | Other Constituents (mM) | Temperature (°C) |
|---|------------------------------------------------------------|----------------------------|------------------------|-----------------|
| 6 | 18.3±1.4                                                   | A 1                        | S 12.5, Fe$^{3+}$ 1    | -               |
| 6 | 15.8±1.1                                                   | A 1                        | S 25, Fe$^{3+}$ 1      | -               |
| 25| 19.1±1.0                                                   | A 1.5                      | S 50, Fe$^{3+}$ 1      | 25              |
| 26| 10.9±1.6*                                                  | A 1                        | S 50, Fe$^{3+}$ 1, NaCl 1 | 15-17.5         |
| 27| 9.7±1.1                                                    | A 1                        | S 30, Fe$^{3+}$ 1      | 22              |
| 27| 11.9±1.8                                                   | A 1                        | S 30, Fe$^{3+}$ 1      | 22              |
| 28| 12.5±1.1                                                   | A 1                        | S 50, Fe$^{3+}$ 1, NaCl 1 | 5               |
| 28| 21.3±0.5                                                   | A 1                        | S 50, Fe$^{3+}$ 1      | 24              |
| 29| 8.2±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5 | 10              |
| 29| 9.1±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, Fo 70 | 20              |
| 29| 10.4±0.1                                                   | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, P 0.6 | 10              |
| 29| 4.4±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, P 0.6 | 10              |
| 29| 0.7±0.1                                                    | G 8                        | S 26, Fe$^{3+}$ 0.2, BE 5, Fo 46 | 20              |
| 29| 1.0±0.1                                                    | G 8                        | S 26, Fe$^{3+}$ 0.2, BE 5, Fo 46, P 0.6 | 20              |
| 29| 4.4±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, XO 0.2 | 10              |
| 29| 6.5±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, BD 0.6 | 10              |
| 29| 6.1±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, Fo 46, XO 0.2 | 20              |
| 29| 6.3±0.0                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5, AC 0.6 | 20              |
| 29| 8.3±0.1                                                    | G 4                        | S 26, Fe$^{3+}$ 0.2, BE 5 | 10              |
| 30| 14±3                                                       | A 1.5                      | S 50, Fe$^{3+}$ 0.5    | 22              |
| 30| 20±5                                                       | A 1.5                      | S 100, Fe$^{3+}$ 0.5   | 22              |
| 30| 22                                                         | A 1.5                      | S 200, Fe$^{3+}$ 0.5   | 22              |
| 30| 11                                                         | A 1.5                      | S 50, XO 0.25          | 22              |
| 30| 5±1                                                        | G 10                       | S 50 & 100, Fe$^{3+}$ 0.5 | 22              |
| 30| 9                                                          | A 1.5, G 3                 | S 50, Fe$^{3+}$ 0.5    | 22              |
| 30| 9                                                          | A 1, G2                    | S 200, Fe$^{3+}$ 0.5, XO 0.2 | 22              |
| 30| 3±1                                                       | A 1.5, G 3                 | S 50 & 100, Fe$^{3+}$ 0.5, XO 0.1 & 0.25 | 22              |
| 31| 14.6±0.1                                                   | G 4                        | S 50, Fe$^{3+}$ 1.5, XO 1.5 | -               |
| 31| 8.1±0.1                                                    | G 4                        | S 50, Fe$^{3+}$ 1.5, XO 1.5 | -               |
| 31| 8.2±0.1                                                    | G + BA                     | S 50, Fe$^{3+}$ 1.5, XO 1.5, BE 5.0 | -               |
| 31| 17.8±0.2                                                   | A 1.5                      | S 50, Fe$^{3+}$ 1.5, XO 1.5 | -               |
| 31| 16.3±0.2                                                   | A 3                        | S 50, Fe$^{3+}$ 1.5, XO 1.5 | -               |
| 32| 1.4                                                        | PVA 20                     | S 50, Fe$^{3+}$ 0.4, XO 0.4 | 20              |

A = agarose, Agar = agar, G = gelatin, PVA = polyvinyl alcohol, S = H$_2$SO$_4$, XO = xylenol orange, BA = benzoic acid, Fo = formaldehyde, P = phenanthroline, AC = acetylacetone, BD = bathophenanthroline disulfonic acid, *Diffusion coefficient calculated in Rae1996
Table 2. Representative selection of R2–dose sensitivities published in the polymer gel dosimetry literature as published in Lepage et al 2001 [17]. Unless otherwise indicated, all polymer gel formulations composed of by weight 3% of the monomers indicated and 5% gelatin or 1% agarose.

| Polymer Gel Dosimeter | Reference | R2–dose sensitivity (S/Gy) | Comment |
|-----------------------|-----------|---------------------------|---------|
| AA, BIS, gelatin      | 33        | 0.259                     | 3-6% monomers |
|                       | 34        | 0.28-0.56                 | 6% monomers |
|                       | 35        | 0.578                     |         |
|                       | 36        | 0.233                     |         |
|                       | 37        | 0.23                      |         |
|                       | 38        | 0.163                     |         |
|                       | 39        | 0.20-0.29                 |         |
|                       | 40        | 0.211                     |         |
| ACA, BIS, gelatin     | 42        | 0.335                     |         |
|                       | 43        | 0.377                     |         |
| VP, BIS, gelatin      | 44        | 0.095                     |         |
| Na methacrylate, BIS, gelatin | 45 | 0.09–0.21              |         |
| MCA, gelatin          | 46        | 0.75–2.66                 |         |
| AA, BIS, agarose      | 47        | 0.28                      |         |

5. PRESAGETM Dosimeters
A new class of polymer dosimeter, PRESAGETM (Heuris Pharma, Skillman, NJ) [24] was proposed in 2003 and based on clear polyurethane combined with leuco-dye leucomalachite green. The components of the dosimeter include an alkyl diisocyanate prepolymer, a hydroxyl reactive polyol along with a catalyst, which polymerises into optically clear polyurethane. Although not suitable for MRI evaluation, the leuco dyes have a maximum absorbance at a wavelength of 633 nm and are therefore suitable for evaluation with a He-Ne laser-based optical scanning system.

Table 3. Different formulations published for normoxic polymer gel dosimeters as published in Senden et al 2001 [22].

| Normoxic dosimeters | Reference | Polymer Gel Dosimeter Formulation |
|---------------------|-----------|----------------------------------|
| MAGIC               | 20        | Methacrylic acid, ascorbic acid, hydroquinone, CuSO4·5H2O, gelatin |
|                     | 48        |                                   |
|                     | 49        |                                   |
| MAGAS               | 49        | Methacrylic acid, ascorbic acid, gelatin |
|                     | 50        |                                   |
| MAGAT               | 49        | Methacrylic acid, tetrakis (hydroxymethyl) phosphonium chloride, gelatin |
|                     | 51        |                                   |
|                     | 52        |                                   |
| nMAG                | 53        | Methacrylic acid, Bis[tetrakis (hydroxymethyl) phosphonium] sulfate, gelatin |
| PAGAS               | 49        | Acrylamide, N,N-methylene-bis-acrylamide, Ascorbic acid, gelatin |
| PAGAT               | 51        | Acrylamide, N,N-methylene-bis-acrylamide, tetrakis (hydroxymethyl) phosphonium chloride, hydroquinone, gelatin |
|                     | 54        |                                   |
|                     | 55        |                                   |
|nPAG                | 53        | Acrylamide, N,N-methylene-bis-acrylamide, Bis[tetrakis (hydroxymethyl) phosphonium] sulfate, gelatin |

6. Evaluation of Gel Dosimeters
Since the work of Gore et al in 1984, the majority of investigations have been undertaken with MRI. However, alternative techniques for evaluation have been introduced including optical computer tomography (CT) in 1996 [56], x-ray CT in 2001 [57], vibrational spectroscopy in 1998 [58] and ultrasound in 2002 [59].

For further information regarding the evaluation of polymer gel dosimeters, the proceedings of the DOSGEL conferences [60,61,62] should be consulted.
7. Other Developments
A significant non-radiotherapy development in gel dosimetry was reported in 2005 by Hill et al [63] in the use of polymer gel dosimeters in measuring the CTDI (Computer Tomography Dose Index) on a diagnostic x-ray CT scanner. This work indicated the potential of using polymer gel dosimeters for diagnostic dose levels.

Another significant non-radiotherapy development in gel dosimetry was research reported by Gore et al in 1997 using polymer gel dosimeters to develop an image quality test tool for MRI [64].

8. DOSGEL Conferences
In June 1995 whilst attending the AAPM in Boston, Clive Baldock and L. John Schreiner decided that it would be appropriate to organise some form of specialist meeting or workshop on gel dosimetry. In September 1996 Clive Baldock and Lars Olsson, whilst attending ESTRO in Vienna, subsequently commenced organising the first international conference on gel dosimetry. This resulted in DOSGEL’99 [60], the first [Fig.1] of the successful DOSGEL conference series and held at the University of Kentucky, USA in 1999. Subsequently DOSGEL 2001 was held at Queensland University of Technology in Brisbane, Australia [Fig.2] and DOSGEL 2004 [62] at the University of Ghent [Fig.3].

9. Conclusion
To date there is still no consensus on optimal gel formulations of gel dosimeters or the optimal evaluation techniques to be used. A result is that this form of 3D dosimetry is yet to be accepted for routine use in the clinic. Until then, there remains much research still to be undertaken.
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