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Diffusion measurement in ferrous infused gel dosimeters

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ABSTRACT

Background: The compositions of Ferrous sulphate, Agarose and Xylenol orange dye (FAX) and Ferrous sulphate, Gelatin and Xylenol orange dye (FGX) in solution of distilled water and sulphuric acid are two tissue-equivalent gel dosimeters. Ionizing radiation causes oxidation of Fe^{2+} ion to Fe^{3+} ions which diffuse through the gel matrix and blur the image of absorbed dose over a period of hours after irradiation.

Materials and methods: 25 mM sulphuric acid, 0.4 mM ferrous ammonium sulphate, 0.2 mM xylenol orange dye and 1% by weight agarose in distilled water named FAX and 0.1 mM ferrous ammonium sulphate, 0.1 mM xylenol orange dye, 50 mM sulphuric acid and 5% by weight gelatin in distilled water named FGX are used as two gel dosimeters. All chemicals were supplied by Sigma Aldridge Company, Germany. The gels were poured in Perspex casts and were irradiated to a beam of X ray from linear accelerators or x ray machine.

Results: In this study diffusion coefficients of FAX and FGX dosimeters have been measured through a computer program for different temperatures. The ferric ion diffusion coefficient (D) for the FAX and FGX dosimeters were measured as $(1.19 \pm 0.03) \times 10^{-2}$ $cm^2.hr^{-1}$ and $(0.83 \pm 0.03) \times 10^{-2}$ $cm^2.hr^{-1}$ respectively at room temperature.

Conclusion: For both dosimeters the diffusion coefficients decreased with gel storage temperatures down to 6°C. FGX dosimeters have advantage of lower diffusion coefficient for a specified temperature. *Iran. J. Radiat. Res.; 2003; 1(2): 79 – 86.*

Keywords: *Chemical dosimeter, gel dosimetry, diffusion, ferrous sulphate gels, fricke gel dosimeter.*

INTRODUCTION

Both experiment and theory have shown that diffusion can result from pressure gradient (pressure diffusion), temperature gradient (thermal diffusion), external force fields (forced diffusion) and concentration gradients. In this study the last type of diffusion is of interest, i.e. the dicussion is limited to diffusion in isothermal, isobaric systems with no external force field gradient. Diffusion coefficient (D) is expressed in square meter per second or cm^2/hr.

Gel dosimeter is a non-steady-state medium in which the diffusion coefficient changes with time (T) as well as distance (X) and concentration (C). The relation between these parameters is expressed in the following equation:

$$C_T(X) = A\left(\frac{1}{4DT\pi}\right)^{1/2} e^{-x^2/4DT}$$

Diffusion coefficient in gels also found to be temperature dependent. Therefore the equation is solved for constant temperature and a computer program is written to accept data at zero time and data for a time (t) hour later, along with a

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pre-selected value of the diffusion coefficient. The program calculated the optical density (proportional to ferric ion concentration) after (t) hours using the diffusion coefficient D with a convolution algorithm. The calculated data was compared to the measured (t) hours data with calculation of their squared difference. Different values of D were input to the program and the value of D for which the squared difference was minimized was accepted as the diffusion coefficient for that specified constant temperature. Many works have been devoted to the study of ferric ion diffusion (Schuls *et al.* 1990, Day 1990, Olsson *et al.* 1992, Balcom *et al.* 1993, 1995, Brunt *et al.* 1994, Harris *et al.* 1996, Rae *et al.* 1996, Kron *et al.* 1997, Pedersn *et al.* 1997, Baldock *et al.* 1997). Researchers have measured the ferric ion diffusion coefficient to predict the effects of diffusion and separate the measurement of dose from diffusion.

One way of combating ferric ion diffusion would be increase the concentration of gelation agent. However gels with agarose more than 1.0% and gelatin more than 9% by weight are difficult to prepare and have increased optical density.

MATERIALS AND METHODS

The FAX gel ingredients are 25 mM sulphuric acid, 0.4 mM ferrous ammonium sulphate, 0.2 mM xylenol orange dye and 1% by weight agarose in distilled water .All chemicals were supplied by Sigma Aldridge Company, Germany. An optimum recipe for the FGX gel was found to be 0.1 mM ferrous ammonium sulphate, 0.1 mM xylenol orange dye, 50 mM sulphuric acid and 5% by weight gelatin in distilled water. Prepared gel was poured in Perspex casts with 1cm thickness and 10 cm x 15 cm width and length respectively. Irradiation was performed with an orthovoltage unit (Toshiba Model KXC-19-2) and three linear accelerators (one Varian Clinac 1800 and two Varian Clinac 600C). The gel dosimeters were scanned for optical density change due to diffusion with a laser scanning system similar to that described by Tarte *et al.* (1996).

In this study, the ferric ion diffusion coefficients were determined for the standard FAX and FGX gel dosimeters. Ferric ion diffusion in the FAX gel dosimeter was also measured for different temperatures of the gel. Two different experimental techniques involving optical scanning of the gels were used. First, ferric ion diffusion was measured across the boundary of two gels, with one gel only containing ferric ions (figure 1). Second, a gel phantom was irradiated and the ferric ion diffusion monitored over a period of hours after irradiation in one dimension (figure 2). Finally, ferric ion diffusion was studied in irradiated gels and monitored for gels irradiated with a HDR brachytherapy source.

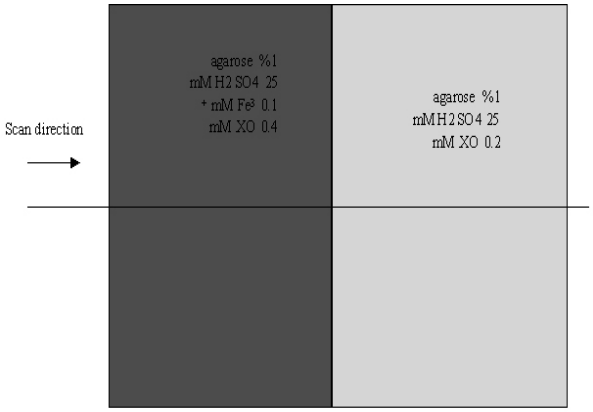

Figure 1. Configuration of FAX gel phantom for diffusion measurement in one dimention across a boundary.

Experimental Procedures for one dimensional diffusion in a gel boundary

A Perspex phantom was half filled with 1% agarose, 25 mM H2SO4 and 0.2 mM XO. When set a gel consisting of 1% agarose, 25 mM H2SO4, 0.4 mM XO and 0.1 mM Ferric ions was poured on top of the pre-set gel to completely fill the phantom. With these procedures a balanced concentration was achieved in both sides of the gel phantom. Since the XO complexes with the ferric ion in a 1:2 ratio, i.e. 0.2 mM of XO in the second part of

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phantom complexes with ferric ions, while the residual 0.2 mM XO balancing the 0.2 mM XO in the first part. In this situation there will be no diffusion of XO from one gel to the other (figure 1).

Experimental procedures for one dimension diffusion in irradiated phantom

As the boundary situation described in previous section, it does not exist for gels prepared normally for irradiation. Diffusion across such a boundary may not be indicative of diffusion throughout a standard gel. In this section two phantoms of FAX gel were made simultaneously from one batch of gel. An area of 12 x 3 cm2 in the middle of one of the phantoms was irradiated (figure 2) and the other was kept as blank. Note that with this irradiation technique, there is a concentration gradient in one dimension only, which causes a change of concentration in this very same dimension.
The irradiated and unirradiated gels were scanned and optical density values obtained from unirradiated gel was subtracted from irradiated values to remove the effect of thermal oxidation. Since the irradiated and unirradiated gels have different rate of thermal oxidation this subtraction is only a rough approximation. Figure 5 shows the result of this experiment.

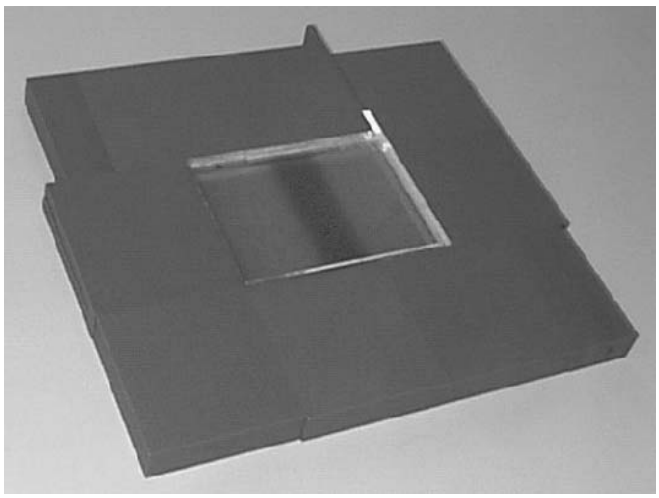

Figure 2 Illustrating the configuration for irradiation of a $10\times15\times1 cm^3$ gel phantom for diffusion measurements.

Experimental procedures for diffusion measurement in an HDR brachytherapy source

A flexible brain implant needle with 2 mm external and 1.5 mm internal diameter was fixed in the middle of the gel phantom and liquid FAX gel was poured into the phantom. The needle was stabilized with setting of the gel. The irradiation time was chosen to give a 10 Gy dose at 10 mm from the source. In HDR brachytherapy source, the outer wall of the needle received a dose of 250 Gy and dropped to 10 Gy at 10 mm distance.

RESULTS

One dimensional diffusion in a gel boundary

The phantom in figure 1 was scanned in different times and a single linear scan perpendicular to the gel boundary was recorded each time. Data (i.e. optical density as a function of position) obtained from scanning 40 minutes after gel preparation was accepted as zero time data (zero diffusion) and scan data for other times were compared to the zero time data (figure 3). Applying the above mentioned computer program and using one set of data obtained from figure 3,(i.e. data belonging to 23.3 hours), a diffusion coefficient of D = 1.27 x 10^{-2} cm^2/hr was applied to the measured data and corrected theoretical optical density values was calculated. Figure 4 shows the measured and calculated data. Diffusion coefficient values ranges from 1.2 x 10^{-2} cm^2/hr to 1.28 x 10^{-2} cm^2/hr with average value of 1.25 x 10^{-2} cm^2/hr (see table 1).

Diffusion in irradiated gels measured in one dimension

Phantom in figure 2 was scanned and the same computer program was used to obtain values of diffusion coefficient from optical density scans at different times after irradiation. Figure 5 is one of the results of such calculation with D value of 1.25 x 10^{-2} cm^2/hr for 490 minutes after irradiation. In this experiment D values ranges from 0.95 x 10^{-2} cm^2/hr to 1.25

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Table 1. Diffusion coefficient D in units of 10^{-2} cm^2/hr for different times in hour at room temperature for FAX and FGX

FAX	Time	1.58	2.58	3.58	4.58	5.58	6.58	7.58	8.58	9.58	22.7	28.8	Average
	D	1.2	1.23	1.24	1.24	1.24	1.25	1.26	1.26	1.27	1.27	1.28	1.25
FGX	Time	1.92	3.42	4.5	5.58	6.58	7.5	8.5	10.3	14.4	18.1	-	Average
	D	0.95	1.10	1.15	1.20	1.20	1.20	1.25	1.25	1.25	1.30	-	1.19

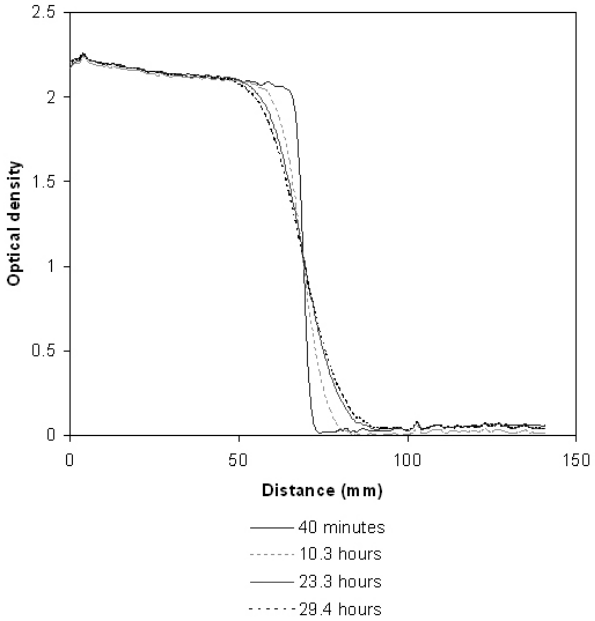

Figure 3. Optical density as a function of position in a gel comprising two sections. One gel section contained 0.4 mM XO and 0.1 mM Fe^{3+} and the other section 0.2 XO. Both sections were 1% by weight agarose and 25 mM H_2SO_4.

x 10^{-2} cm^2/hr with average value of 1.19 x 10^{-2} cm^2/hr (table 1). It seems that lower value of averaged diffusion coefficients in the second experiment is due to removal of thermal oxidation.

Diffusion measurements at different temperatures

Thus far measurement was done in the room temperature. There may be advantages in storing, irradiating and scanning the gels at temperatures lower than room temperature. At temperatures lower than room temperature, thermal oxidation would be reduced and the diffusion coefficient would be expected to be lower.

In this section, the diffusion coefficients in FAX gels were measured for refrigerator temperature (6°C), cold-water temperature (15°C) and normal water temperature (20°C). A temperature controller unit and stirrer were used with the gel in a water bath to keep the water temperature constant. Scanning and diffusion coefficient measurement procedures were the same as the previous sections. Table 2 is result of these experiments.

As it can be seen from table 2, for each temperature, diffusion coefficient is increasing with time and it is lower for temperatures below room temperature.

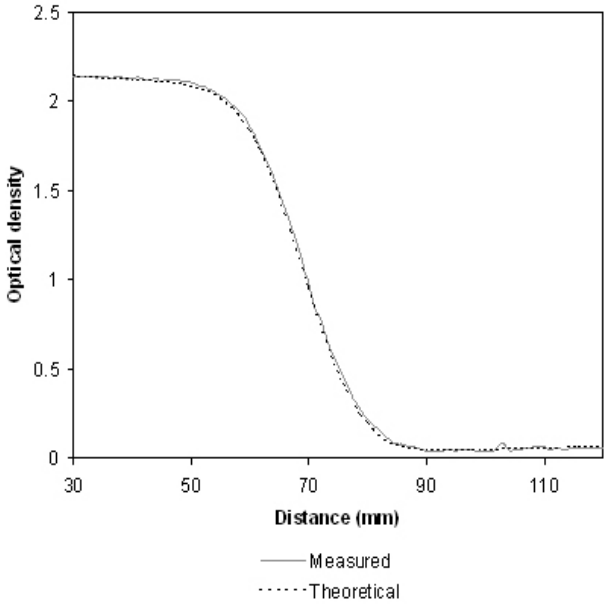

Figure 4. Measured and theoretical optical density profile after 23.3 hours.
Diffusion Coefficient: D = 1.27 × 10^{-2} cm^2/hr.

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Table 2. Diffusion coefficients D in unit of 10^{-2} cm^2/hr for the FAX gel dosimeter at diffusion temperatures for different time (t) after first scan (hr).

Refrig. (6 ± 2)°C		Water (15 ± 2) °C		Water (20 ± 2) °C		Room (23 ± 1) °C	
Time(h)	D	Time(h)	D	Time(h)	D	Time(h)	D
8.25	1.05	1.42	0.90	1.42	1.30	1.92	0.95
9.83	1.05	3.17	1.05	3.33	0.90	3.42	1.10
12.00	1.0	4.17	1.00	4.25	1.00	4.5	1.15
15.33	1.05	5.33	1.10	5.42	0.90	5.58	1.20
17.08	1.20	8.5	1.05	8.58	1.25	6.58	1.20
-	-	9.83	1.05	9.5	1.15	7.50	1.20
-	-	12.33	1.00	11.33	1.20	8.50	1.25
-	-	15.58	1.10	12.58	1.15	10.33	1.25
-	-	17.17	1.10	13.67	1.30	14.42	1.25
-	-	-	-	24.58	1.35	18.08	1.30
Average	1.070	-	1.039	-	1.150	-	1.185
STDEV	0.068	-	0.065	-	0.165	-	0.100
STD error	0.03	-	0.02	-	0.05	-	0.03

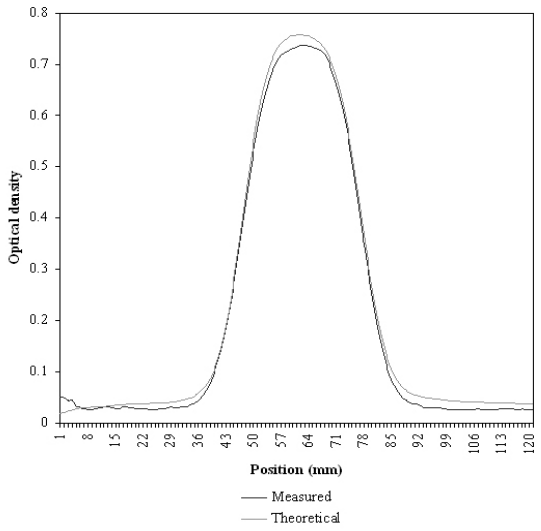

Figure 5. Calculated and measured optical density after 490 minutes for the FAX gel dosimeter at room temperature.

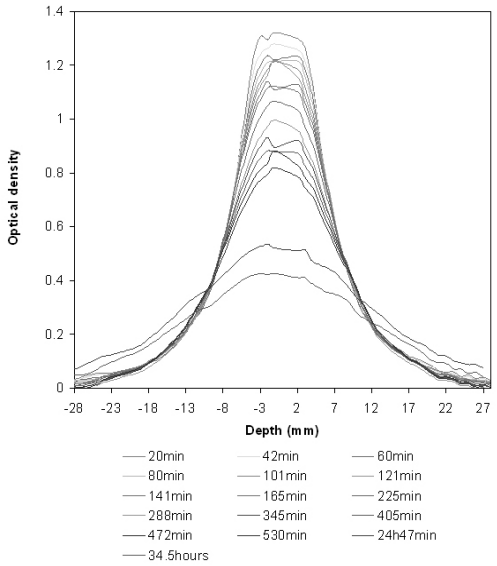

Figure 6. Optical density versus depth for a FAX gel dosimeter irradiated with a HDR Ir^{192} source. 10 Gy dose is delivered at 10 mm from centre of the source. Thickness of the FAX gel is 5 mm.

Diffusion in a FAX gel irradiated with a HDR brachytherapy source

Data from this experiment is shown in figure 6. According to data obtained from this experiment, the optical density decreased by 10% within the first 20 minutes between the first and the second Scan. As the time elapses, the curves representing optical density as a function of position broadened. The optical density adjacent to the source decreased with time and a third order polynomial fit the data with R2 = 0.99, (figure 7). This graph shows that 18 hours after irradiation, the optical density reaches an approximate constant value and changes very slowly with time after that.

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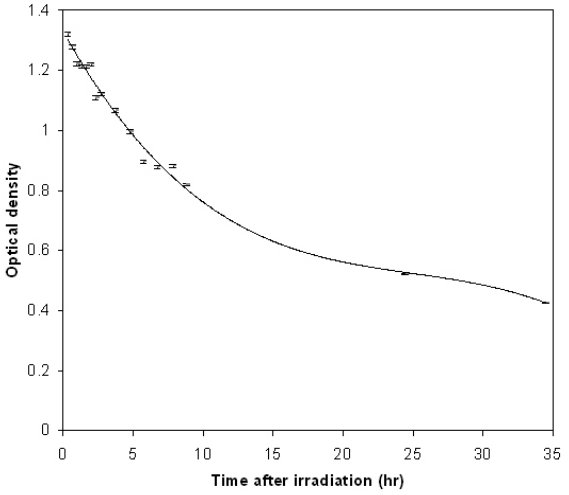

Figure 7. Optical density versus time at a distance of 1 mmfrom an Ir^{192} HDR source in a FAX gel section.

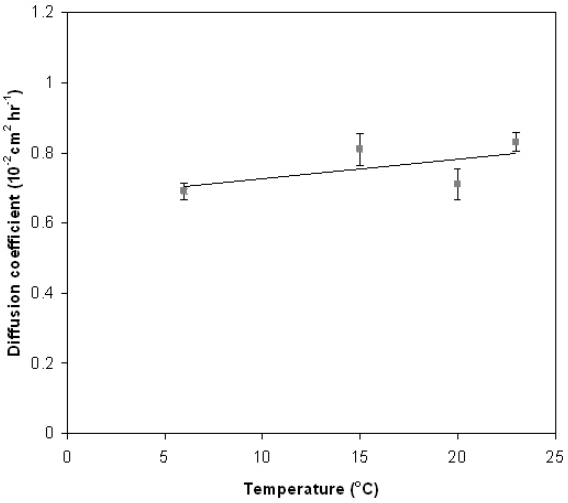

Figure 9. Diffusion coefficient versus temperature for the FGX dosimeter.

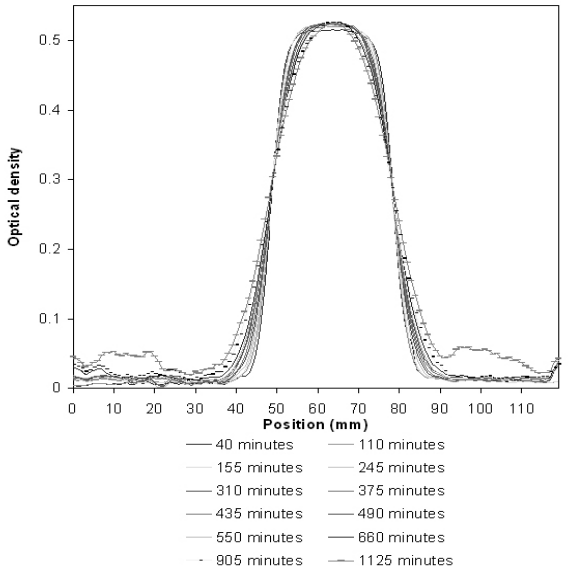

Figure 8. Optical density versus position for different times after irradiation of the FAX gel dosimeter at room temperature.

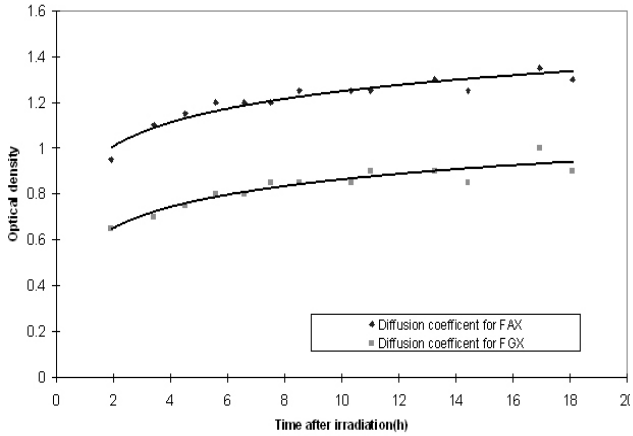

Figure 10. Calculated diffusion coefficients versus time for the FAX and GFX at 23ºC (air conditioned room).

Diffusion in the FGX in one dimension for different temperatures

Experimental procedures for FGX dosimeter are the same as procedures described for FAX dosimeter in dimensional irradiated phantom. In order to compare the behavior of FAX and FGX gels, both dosimeters were prepared, stored, scanned and irradiated under the same experimental conditions. The variation of optical density due to diffusion versus position in FGX dosimeter is shown in figure 8. As it can be observed in the graph, the curves are broadening with time that is indicating the increase of diffusion coefficient in FGX with time for a constant temperature. The diffusion coefficient in FGX also tends to increase with temperature as it can be seen from figure 9.

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DISCUSSION

The ferric ion diffusion coefficient (D) for the FAX and FGX dosimeters were measured as $(1.19 \pm 0.03) \times 10^{-2}$ cm^2/hr and $(0.83 \pm 0.03) \times 10^{-2}$ cm^2/hr respectively at room temperature. For both dosimeters the diffusion coefficients decreased with gel storage temperatures down to 6^{o}C. Comparison of optical density values indicate that optical density in FAX gel dosimeter is relatively higher than FGX dosimeter (figure 10); this means FGX dosimeters have advantage over the FAX ones.

The values of diffusion coefficients in FGX dosimeter is also lower than that of FAX and it again emphasizes the superiority of FGX dosimeters if a gel dosimeter is to be used for a longer period of time.

Measurement of diffusion coefficient for the FGX dosimeter is reported by Rae *et al.* (1996). They measured $D = 0.44 \times 10^{-2}$ cm^2/hr for a gel containing 0.2 mM xylenol orange, 26 mM sulphuric acid and 4% by weight gelatin at 10^{o}C. The higher value of diffusion coefficient in this work in comparison to the value from Rae *et al.* could be due to higher temperature (23^{o}C as opposed to 10^{o}C) and higher sulphuric acid concentration (50 mM as opposed to 26 mM). Sulphuric acid weakens the gel structure, thus a gel with a higher sulphuric acid concentration might be expected to allow faster ferric ion diffusion.

Diffusion coefficient obtained for FAX gel dosimeter in this study is lower than the value found by Kron *et al.* (1997) who measured $D = 1.21 \times 10^{-2}$ cm^2/hr for a gel containing 1.5% agarose, 50 mM sulphuric acid and 0.25 mM xylenol orange. It is also lower than the values found by Olsson *et al.* (1992), Schulz *et al.* (1990), Baldock *et al.* (1995a) and Harris *et al.* (1996) who found the values of 1.91×10^{-2} cm^2/hr, 1.58×10^{-2} cm^2/hr, 1.25×10^{-2} cm^2/hr $\pm$ 0.09 and 2.08×10^{-2} cm^2/hr respectively.

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