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Fricke gel diffusion coefficient measurements for applications in radiotherapy level dosimetry

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HIGHLIGHTS

- FXG investigations in the radiotherapy.
- Gaussian and ISQR methodologies are tool of determined diffusion coefficient corrected.
- The FXG diffusion coefficient increases with the storage temperature.

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ABSTRACT

In gel dosimetry applied to radiotherapy, the space–time corrections are necessary due to the diffusion of ions in the oxidized solution dosimetry. Consequently, methodologies are applied in order to determine diffusion coefficients corrected in space and time. Therefore, in this study the dosimetric solution Fricke Xylenol Gel (FXG) was modified and applied to two Gaussian and ISQR methodologies for comparison of the diffusion coefficients obtained. The results show that the FXG system can be modified for new applications in radiotherapy, and it may be corrected in space–time to the appropriate methodologies in the determination of diffusion coefficients.

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1. Introduction

The standard Fricke solution for radiation dosimetry has been modified over the years: with the addition of xylenol orange (XO) dye and 300 Bloom gelatin, a solution becomes more sensitive and the doses of ionizing radiation more stable (Bero et al., 1999, 2000, 2001; Davies and Baldock, 2008, 2010). Due to these modifications, the gels have obtained dosimetric applications in radiotherapy as: beam control and planning of patients in the treatment of cancers. The Fricke Xylenol Gel (FXG) dosimeter is based on the Fe^{2+} to Fe^{3+} oxidation due to ionizing radiation, forming the $[\text{Fe}^{3+}\text{-OX}]$ complex, whose absorbance peak is centered at 585 nm, with similar characteristics to those of soft tissue, such as density and effective atomic number, linear dependence with the absorbed dose in a broad range used in radiotherapy (Alva-Sánchez et al., 2014; Pirani et al., 2013). Although the gels were added to stabilize the signal, even oxidized ions present a certain freedom to diffuse potentially, affecting readings into significant post-irradiation time (Davies et al., 2013; Maeyama et al., 2014; Oliveira et al., 2007, 2009a, 2009b). Recently, FXG has been used in studies of physical parameters in radiotherapy, dosimetry

of small fields and the development of equipment for reading dosimetric systems (Calcina et al., 2007; Sampaio et al., 2013a, 2013b). New formulations from the dosimeter were also accomplished, making it applicable to low doses of radiation, such as those used in diagnostic radiology procedures (Pirani et al., 2009).

Studies were undertaken in an attempt to reduce the diffusion coefficient (D) of the oxidized ions of interest, or by changing their reagents, masses or concentrations; the results were significant which demonstrates the importance of forming new dosimetric gels (Abukassem and Bero, 2010; Balcom et al., 1995; Kron et al., 1997).

The objective of this research is the investigation of methodologies to obtain diffusion coefficients associated with the variation of their reagents and stored temperature. This study may be useful in choosing dosimetric gels for radiotherapy procedures with smaller diffusion coefficients of measurements in space–time.

2. Materials and methods

In this work, the variation in the formulation of FXG in terms of its reagents was evaluated, with four new solutions compared with the FXG in terms of diffusion coefficients.

The FXG components changed were as follows: 300 Bloom gelatin, weight of the mass by the total weight of the solution (w/w), was

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changed from 5% to 6% and 8%, the dye concentration from 0.10 mM to 0.12 mM and 0.15 mM, maintaining fixed the other concentrations of FXG (5% w/w+0.10 mM) and obtaining four different solutions: A=(6% w/w+0.10 mM), B=(8% w/w+0.10 mM), C=(5%+0.12 mM), and D=(5% w/w+0.15 mM). After preparing the FXG solution and the A, B, C, and D solutions, they were inserted into standard cuvettes with dimensions of $1.25 \times 1.25 \times 4.5 \text{ cm}^3$.

All irradiations were performed with a VARIAN® CLINAC 6/100 linear accelerator, at a 100 cm of source–surface distance (SSD), 6 MV photon beam and $2000 \text{ mGy min}^{-1}$ of dose rate, at the maximum depth equal to 1.5 cm and the beam is uniform across the surface of the samples. A PMMA plate was used to provide electronic equilibrium and 10 plates with thickness of 1 cm were positioned under the cuvettes to avoid backscattering contributions.

For the measurements of the calibration curve, the cuvettes were used for each irradiation procedure, the FXG dosimeter cuvettes together with the FXG with new concentrations (Fig. 1). The selected absorbed doses were 1, 3, 5, 7 and 10 Gy of radiation; for each beam three cuvettes of each dosimeter were selected to obtain the average of their readings in the spectrophotometer Femto-800 XL.

For the determination of the diffusion coefficients, the field size was half-blocked and irradiated with an absorbed dose of 5 Gy (Fig. 2). In this case, eight cuvettes were irradiated, four with regular FXG and the other four with the modified concentrations of FXG_{XO} (5% w/w+0.12 mM). After the irradiation, the cuvettes were stored at the temperatures of 13 °C and 25 °C.

In this work, two regions of interest, the irradiated portion and the non-irradiated portion of the cuvettes containing FXG solutions, were studied as a semi-infinite system together at $t=0$. The associated Fick equation provided the diffusion coefficients (Oliveira et al., 2009a, 2009b). These diffusion coefficients (D) can then be determined by applying these two configuration methodologies: Inverse Square Root Function Method (ISQR) and Gaussian Function (Kron et al., 1997). For the first time, a comparison of these two approaches will be applied to the FXG dosimeter, using experimental data for the irradiated region using the methodology associated with the ISQR step function; in this case, the curvature is proportional to the calculated absorbed dose D . For the Gaussian methodology, the derivative of the step function is used, and it associates the half-width of the curve for determination of absorbed dose D . According to the Gaussian methodology, the depen-

dence over time provides the following meanings: results for small times not as accurate as the ions encounter barriers on their way; with longer times results from the methodology Gaussian approach for the results obtained by ISQR methodology, for longer times the Fe^{+3} ions have greater opportunity to explore the size of the cuvettes; the time for these measures should be informed and not mixed; there will not be result without; there is a limitation in that Gravitational settling is neglected; a ramification of this methodology would be the application of diffusion in isotropic and anisotropic media, differentiating between irradiated and non-irradiated parts. For the purpose of this research, the two methodologies can be applied according to the measurements to be carried out in radiotherapy for post-irradiation. Measurements in which the answer should be small times. It is advisable to use Gaussian methodology for a large time step in the ISQR methodology which is preferable.

3. Results and discussion

Fig. 3 shows the dose–response curves of the FXG solutions in function of the absorbed dose, and the sensitivities found for 5%, 6%

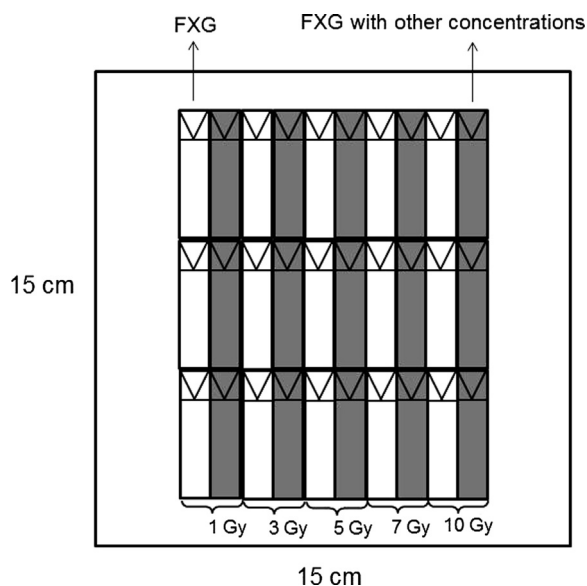


Fig. 1. Experimental set-up arrangement for measuring the calibration curve of dosimeters and FXG and FXG with other concentrations.

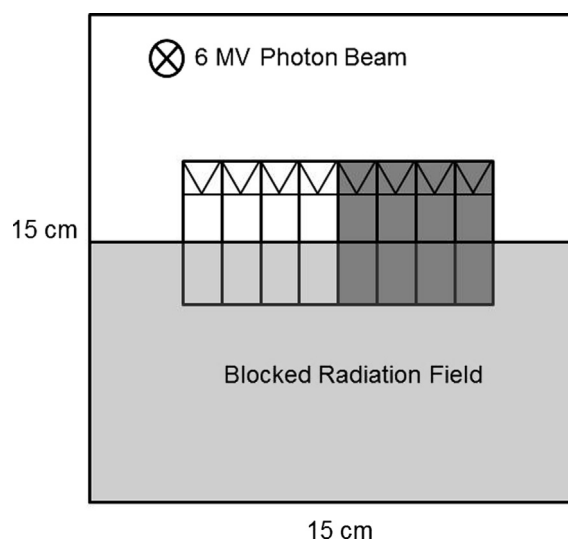


Fig. 2. Irradiations with 6 MV photons and radiation field blocked for FXG cuvettes.

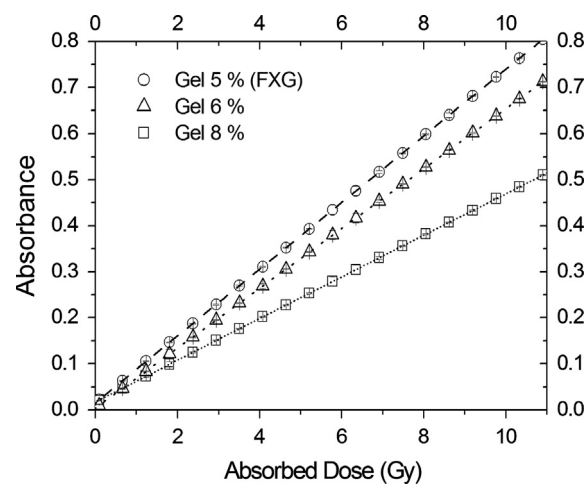


Fig. 3. Dose–response curves for different concentrations of 300 Bloom gelatin in the FXG solutions for absorbed doses up to 10 Gy. The concentrations used were 5% gel (FXG=5% w/w+ 0.10 mM), 6% gel (A=6% w/w+ 0.10 mM) and gel 8% (B=8% w/w+ 0.10 mM).

and 8% w/w were respectively 0.072, 0.065 and 0.045 Gy⁻¹. A linear behavior was obtained in all cases; it may be observed that an increase in the gelatin concentration of 6–8% w/w caused a 30% decrease in the response sensitivity; this fact may be due to the effect of pH variation in the mixed solution; this change causes the gel not to be so clear as standard, thus modifying the sensitivity.

In Fig. 4, the dose–response curves of xylenol FXG solutions present linearity in all curves of different concentrations of FXG; the change in xylenol concentrations associated a percentual increase in its sensitivity of approximately 24% to 0.15 mM xylenol. This fact can be explained by the high concentration of XO and consequently the formation of a greater complex Fe⁺³-OX in the FXG solution.

The diffusion coefficients obtained for the FXG regular and modified solutions using the Gaussian and ISQR methodologies are presented in Table 1. Independently on the FXG solution irradiated, the diffusion coefficients increase with the storage temperature and the absence of xylenol. Based on the results of Table 1, it can be concluded that the Gaussian method is time dependent, and it presents decreasing values with the post-irradiation time. For the ISQR methodology, the values are constant for each irradiated solution, which are time-independent. After a certain time *t*, the post-irradiation values obtained by the Gaussian method tends to ISQR values; this fact may be essential for the application of these two correction methodologies in spatial and temporal measurements in radiotherapy, using 3D dosimetric gels.

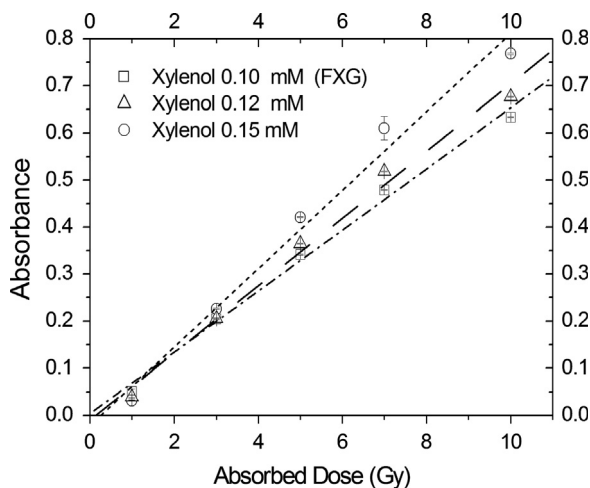


Fig. 4. Dose–response curves for different concentrations of xylenol in the FXG solutions for absorbed doses up to 10 Gy. The concentrations used were xylenol 0.10 mM (FXG=5% w/w+0.10 mM), xylenol 0.12 mM (C=5% w/w+0.12 mM) and xylenol 0.15 mM (D=5% w/w+0.15 mM).

Table 1

Diffusion coefficients for the FXG and FXG_{XO} using the ISQR and Gaussian methodologies. The concentrations used were 5% w/w+0.10 mM for FXG (13 °C) and FXG (25 °C); 5% w/w+0.12 mM for FXG_{XO} (13 °C) and FXG_{XO} (25 °C).

Time (h) Post-irradiation	Diffusion coefficient by the Gaussian method (mm ² /h)			
	FXG (13 °C)	FXG _{XO} (13 °C)	FXG (25 °C)	FXG _{XO} (25 °C)
1	0.86 ± 0.02	0.74 ± 0.02	1.01 ± 0.01	0.75 ± 0.02
2	0.54 ± 0.03	0.50 ± 0.02	0.90 ± 0.02	0.67 ± 0.01
3	0.37 ± 0.01	0.33 ± 0.01	0.84 ± 0.02	0.46 ± 0.01
4	0.25 ± 0.01	0.22 ± 0.01	0.63 ± 0.01	0.35 ± 0.02
5	0.22 ± 0.02	0.17 ± 0.01	0.58 ± 0.01	0.26 ± 0.01
7	0.14 ± 0.02	0.13 ± 0.01	0.57 ± 0.01	0.24 ± 0.01
	Diffusion coefficient by the ISQR method (mm ² /h)			
	0.13 ± 0.01	0.11 ± 0.01	0.57 ± 0.01	0.24 ± 0.01

4. Conclusions

From the results, it can be concluded that: 1) the increase in the concentration of 300 Bloom gelatin decreases the gel response sensitivity; 2) the increase in the concentration of xylenol increases the response sensitivity of the dosimetric gel; 3) the FXG diffusion coefficient increases with the storage temperature; 4) different results for the diffusion coefficients were found for FXG, one time-independent and other time-dependent, respectively for ISQR and Gaussian methodologies; 5) it is expected that the diffusion coefficient for the chemical dosimeters is from 0.3 to 2 mm²/h, this being associated with a change in temperature; the results obtained for the two methods, Gaussian and ISQR, for hot temperature are within this range; the value of diffusion coefficient for the cold temperature is lower for the established limit, which provides greater stability FXG dosimetry information. This fact demonstrates that the coefficient of diffusion for FXG can be determined with the two research methodologies, and the values found agree with the results obtained through other methods (Balcom et al., 1995; Gambarini et al., 2004; Kron et al., 1997; Pedersen et al., 1997; Tseng et al., 2005). This work shows that the FXG dosimeter can be modified for new applications in radiotherapy, and it may be corrected in space–time according to the appropriate diffusion coefficients.

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References

- Abukassem, I., Bero, M.A., 2010. Application of radiochromic gel detector (FXG) for UVA dose measurements. *Radiat. Phys. Chem.* 79, 1209–1214.
- Alva-Sánchez, M.S., Oliveira, L.N., Petchevist, P.C., Moreira, M.V., Almeida, A., 2014. Beta planar source quality assurance with the Fricke xylenol gel dosimeter. *Radiat. Phys. Chem.* 96, 56–59.
- Balcom, B.J., Lees, T.J., Sharp, A.R., Kulkarni, N.S., Wagner, G.S., 1995. Diffusion in Fe (II/III) radiation dosimetry gels measured by magnetic resonance imaging. *Phys. Med. Biol.* 40, 1665–1676.
- Bero, M.A., Gilboy, W.B., Glover, P.M., Keddie, J.L., 1999. Three-dimensional radiation dose measurements with ferrous benzoic acid xylenol orange in gelatin gel and optical absorption tomography. *Nucl. Instrum. Methods Phys. B* 422, 617–620.
- Bero, M.A., Gilboy, W.B., Glover, P.M., El-Masri, H.M., 2000. Tissue-equivalent gel for non-invasive spatial radiation dose measurements. *Nucl. Instrum. Methods Phys. B* 166, 820–825.
- Bero, M.A., Gilboy, W.B., Glover, P.M., 2001. Radiochromic gel dosimeter for three-dimensional dosimetry. *Radiat. Phys. Chem.* 61, 433–435.
- Calcina, C.S.G., Oliveira, L.N., Almeida, C.E., Almeida, A., 2007. Dosimetric parameters for small field sizes using Fricke xylenol gel, thermoluminescent, film dosimeters, and an ionization chamber. *Phys. Med. Biol.* 52, 1431–1439.
- Davies, J.B., Bosi, S.G., Baldock, C., 2013. Dosimetry aspects of a non-diffusing genipin–gelatin gel. *Radiat. Phys. Chem.* 83, 19–27.
- Davies, J.B., Baldock, C., 2010. Temperature dependence on the dose response of the Fricke–gelatin–xylenol orange gel dosimeter. *Radiat. Phys. Chem.* 79, 660–662.
- Davies, J.B., Baldock, C., 2008. Sensitivity and stability of the Fricke–gelatin–xylenol orange gel dosimeter. *Radiat. Phys. Chem.* 77, 690–696.
- Gambarini, G., Birattaria, C., Mariani, M., Marchesini, R., Pirola, L., Prestini, P., Sella, M., Tomatis, S., 2004. Study of light transmittance from layers of Fricke–xylenol–orange–gel dosimeters. *Nucl. Instrum. Methods Phys. Res. B* 213, 321–324.
- Kron, T.D., Jonas, D., Pope, J.M., 1997. Fast T1 imaging of dual gel samples for diffusion measurements in NMR dosimetry gels. *Magn. Reson. Imaging* 15, 211–221.
- Maeyama, T., Fukunishi, N., Ishikawa, K.L., Furuta, T., Fukasaku, K., Takagi, S., Noda, S., Himeno, R., Fukuda, S., 2014. A diffusion-free and linear-energy-transfer-independent nanocomposite Fricke gel dosimeter. *Radiat. Phys. Chem.* 96, 92–96.
- Oliveira, L.N., Calcina, C.S.G., Parada, M.A., Almeida, C.E., Almeida, A., 2007. Ferrous xylenol gel measurements for 6 and 10 MV photons in small field sizes. *Braz. J. Phys.* 27, 1141–1146.

- Oliveira, L.N., Zimmerman, R.L., Moreira, M.V., Ila, D., Almeida, A., 2009a. Determination of diffusion coefficient in Fricke xylenol gel dosimeter after electron beam bombardment. *Surf. Coat. Tech.* 203, 2367–2369.
- Oliveira, L.N., Calcina, C.S.G., Parada, M.A., Almeida, C.E., Almeida, A., 2009b. 6 MV wedge photon beam profiles with the Fricke xylenol gel dosimeter. *Braz. J. Phys.* 39, 615–618.
- Pedersen, T.V., Olsen, D.R., Skretting, A., 1997. Measurement of ferric diffusion coefficient in agarose and gelatine gels by utilization of the evolution of a radiation induced edge as reflected in relaxation rate images. *Phys. Med. Biol.* 42, 1575–1585.
- Pirani, L.F., Moreira, M.V., Costa, J.J.L., Oliveira, L.N., Caldas, L.V.E., Almeida, A., 2013. Fricke dosimeter gel measurements of the profiles of shielded fields. *Appl. Radiat. Isot.* 82, 239–241.
- Pirani, L.F., Oliveira, L.N., Petchevist, P.C.D., Moreira, M.V., Ila, D., Almeida, A., 2009. New chemical Fricke gel radiation dosimeter. *J. Radioanal. Nucl. Chem.* 280, 259–262.
- Sampaio, F.G.A., Oliveira, L.N., Moreira, M.V., Petchevist, P.C.D., Almeida, C.E., Almeida, A., 2013a. 8 and 10 MeV electron beams small field-size dosimetric parameters through the Fricke xylenol gel dosimeter. *IEEE Trans. Nucl. Sci.* 60, 572–577.
- Sampaio, F.G.A., Del Lama, L.S., Sato, R., de Oliveira, D.M.M., Czelusniak, C., Oliveira, L.N., Almeida, A., 2013b. Quality assurance of a two-dimensional CCD detector system applied in dosimetry. *IEEE Trans. Nucl. Sci.* 60, 810–816.
- Tseng, Y.J., Huang, S.C., Chu, W.C., 2005. A least-squares error minimization approach in the determination of ferric ion diffusion coefficient of Fricke-infused dosimeter gels. *Med. Phys.* 32, 1017–1023.