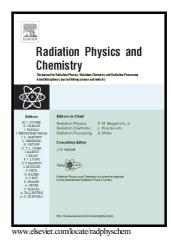
Author's Accepted Manuscript

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 PII:
 S0969-806X(17)31025-3

 DOI:
 https://doi.org/10.1016/j.radphyschem.2018.04.031

 Reference:
 RPC7838

To appear in: Radiation Physics and Chemistry

Received date: 2 October 2017 Revised date: 23 April 2018 Accepted date: 26 April 2018

Cite this article as: Anderson Vinicius Silva Alves, Wandson Santos de Almeida, Eliana Midori Sussuchi, Luigi Lazzeri, Francesco d'Errico and Susana O. de Souza, Investigation of chelating agents/ligands for Fricke gel dosimeters, *Radiation Physics and Chemistry*, https://doi.org/10.1016/j.radphyschem.2018.04.031

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ACCEPTED MANUSCRIPT

Investigation of chelating agents/ligands for Fricke gel dosimeters

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ABSTRACT

The Fricke solution is an absolute dosimeter which can be combined with some metal-binding organic ligands and thus show measured radiation doses through visible light absorption effects. Adding the solution to a gel matrix also allows mapping doses three-dimensionally, and is known as Fricke-gel dosimetry. The latter has some drawbacks that hinder its practical usage for 3D mapping, namely signal diffusion effects and spontaneous color changes due to auto-oxidation. Ligands play a major role in this respect and xylenol orange has been used extensively. Several alternative ligands and their interactions with Fricke solutions and gels were investigated in this study. Among these ligands, calcichrome (CH), eriochrome blue (EB), methylthymol blue (MTB), and naphthol green (NG) showed the required differentiation between optical absorbance of ferrous vs ferric ions. While our study confirmed the overall advantages of using xylenol orange, it also suggests that methylthymol blue offers potential worthy of further investigation.

Keywords: 3D dosimetry; Ligands; Fricke solution; Fricke gel; Radiotherapy.

1. Introduction

"Fricke solutions" have been used for ionizing radiation dosimetry since 1927 (Fricke and Morse, 1927). The system consists of an acidic solution of Fe^{2+} ions, which oxidize to Fe^{3+} upon irradiation. Appleby *et al.* (1987) suggested infusing this solution in a gel matrix to obtain spatial dose maps by means of magnetic resonance imaging (MRI). Fricke-gels (FG) have been amply investigated (Gore and Kang, 1984; Gambarini *et al.*, 1994 and 2017; Luciani *et al.*, 1996, Chu *et al.*, 2000; Smith *et al.*, 2015; d'Errico *et al.*, 2017; Marini *et al.*, 2017). These gels are considered the most promising 3D dosimeters, however, they suffer from two main drawbacks: the relatively fast diffusion of radiation-induced Fe^{3+} ions and auto-oxidation of Fe^{2+} ions during storage (Penev and Mequanint, 2013).

Fast optical scanning techniques minimize the impact of the ion diffusion effects (Schreiner, 2004; Jordan, 2010), however, it would be preferable to lessen these drawbacks. The use of ligands that bind to the iron ions may help in this respect, by diminishing the diffusion of the Fe^{3+} ions and the auto-oxidation of Fe^{2+} ions. Generally, large ligand molecules that preferentially bind with Fe^{3+} , such as xylenol orange (XO), reduce the diffusion coefficients. On the other hand, ligands that mainly

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