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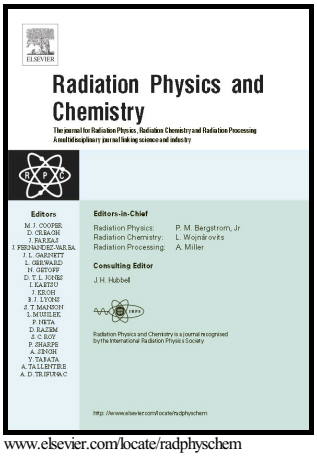

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The role of hydrogels in the radical production of the Fricke-gel-dosimeter

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Abstract — The radiolysis mechanism of the Fricke-gel-dosimeters has been investigated in order to evaluate the role of hydrogels in the radical production. For this purpose, electron paramagnetic resonance (EPR) spectra were acquired for samples frozen and irradiated at 77 K. The analysis was performed by increasing stepwise the temperature and acquiring the EPR spectra at 120 K in order to follow the radical reaction mechanism. The comparison between aqueous- and gel- dosimeters were performed. Both gelatin from porcine skin and PVA (polyvinyl alcohol) were investigated as gel matrix.

Different radical species were identified and qualitatively compared. For gel matrix, peroxyl radicals, stemming from the hydrogel, play an important role in the survival of radicals at higher temperature. Moreover, the Fe^{3+} EPR signal has been studied and compared with the radicals concentration. From this comparison, it is evident the increase of Fe^{3+} concentration is shifted toward higher temperatures with respect to the radical decay. To explain this phenomenon, the intervention of EPR silent species like peroxides is supposed.

Keywords — EPR spectroscopy; Fricke-gel dosimeter; Radical chemistry; Radiation dosimetry.

1. Introduction

The Fricke standard aqueous-dosimeter is an acidic solution of ferrous ions, Fe^{2+}. Upon irradiation, the products of water radiolysis cause the dose-dependent oxidation of ferrous ions into ferric ions, Fe^{3+}. A chelant, the Xylenol Orange (XO), is typically added in the dosimeter formulation to allow an optical measurement in the visible range. In details, the XO forms complexes with the ferric ions which mainly absorb at about 585 nm [Appleby A and Leghrouz A, 1991]. The spatial information of the absorbed dose is preserved by adding a gelling agent, viz. gelatin from porcine skin, agarose and polyvinyl alcohol (PVA) [Schreiner L J, 2004]. Olsson *et al.* reported an increase of the sensitivity of the gel-dosimeter due to a chain reaction in the agarose [Olsson L E *et al.*, 1991]. Since the addition of organic compounds (e.g. sucrose) enhances the radiolytic yield of Fe^{3+}, a similar effect is expected in the case of gelatin and PVA [Healy B J *et al.*, 2003].

As the mass fraction of the gel component is usually less than 5%, the primary effect is the radiolysis of water. Therefore, the organic species stemming from hydrogels might play a key role in the reaction mechanism for the oxidation of iron. In this framework, the radicals are the most important species. The Electron Paramagnetic Resonance (EPR) technique is widely used for a qualitative and quantitative investigation of radicals. Thus, a systematic EPR analysis on both gelatin and PVA matrices was conducted in order to investigate the production and reaction of radicals with respect to the increase of Fe^{3+} concentration.

The radical reaction mechanism occurring at ambient temperature, i.e. in operative conditions, could be investigated by means of pulse radiolysis [Khaikin, G.I. *et al.*, 1996]. As an alternative, a previously developed methodology [Dondi D *et al.*, 2012] was applied using conventional EPR techniques. In details, samples were quickly frozen, irradiated and maintained at 77 K. Subsequently, samples were analyzed in the EPR cavity at 120 K. The temperature was increased stepwise to allow the reactions between radicals and then quickly restored at 120 K for the measurement. Details of the adopted methodology and the obtained experimental results are hereafter discussed.