PRELIMINARY MAGNETIC RESONANCE RELAXOMETRIC ANALYSIS OF FRICKE GEL DOSIMETERS PRODUCED WITH POLYVINYL ALCOHOL AND GLUTARALDEHYDE

by

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Scientific paper
http://doi.org/10.2298/NTRP1703242G

This work describes the preliminary analysis of Fricke gels dosimeters characterized by a new formulation making use of a matrix of polyvinyl alcohol cross-linked by adding glutaraldehyde and analyzed by means of nuclear magnetic resonance relaxometry. In previous optical studies, these gels have shown promising dosimetric features in terms of photon sensitivity and low diffusion of ferric ions produced after irradiation.
In this work, we used a portable nuclear magnetic resonance relaxometer to measure the relaxation times (which are important for dosimetric applications) of these gel materials. For this purpose, we performed a study for optimizing the acquisition parameters with a nuclear magnetic resonance relaxometer. Gel samples were exposed to clinical 6 MV photons in the dose range between 0 and 20 Gy. Nuclear magnetic resonance relaxometry measurements were performed and the sensitivity to photon beams was measured for various values of the Fe^{2+} ion concentration. The analyses pointed out that the MR signal increases as the Fe^{2+} content increases and the increase is about 75 % when the concentration of Fe^{2+} ions is increased from 0.5 mM to 2.5 mM. Furthermore, the sensitivity improvement achieved with increasing the Fe^{2+} concentration is about 60 %. This paper shows that the portable nuclear magnetic resonance relaxometer used for analysis of porous materials can be used for characterization of these dosimetric gels and this study can be considered as the first step for the characterization of these dosimeters which in future could be used for 3-D dose mapping in clinical applications.

Key words: dosimetry, gel, nuclear magnetic resonance, polyvinyl alcohol, glutaraldehyde, radiotherapy

INTRODUCTION

The Fricke gel dosimeters have been long studied because of the dosimetric potential of a solution of ferrous sulfate (for which the radiation-induced production of ferric ions from ferrous ions depends on the irradiation dose) and the rigidity of the gel matrix [1], which hinders the ferric ions from freely diffusing in the solution. This is an advantage with respect to an aqueous solution because the latter does not preserve spatial information on the dose-dependent changes of the local magnetic properties related to the local ferric ions concentration. The two above-mentioned features have made Fricke gel dosimeters ideal candidates for performing 3-D reconstruction of dose distribution, particularly useful in the planning phase of radiation treatments. The great advantage of the Fricke gels is that because of the magnetic properties of their constituents they may be analyzed through magnetic resonance imaging (MRI) and with a single acquisition can provide an image of the 3-D distribution of the radiation-induced ferric ions, and then the corresponding distribution of the dose delivered [2].

A radiation-sensitive device able to map a 3-D distribution of the dose delivered in a clinical setting must combine suitable dosimetric features with the ability to

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capture and store the information on local variations induced by the delivered dose. Dosimetric materials of different types have been studied over the decades to assess their dosimetric performance for various radiation beams. Among the various dosimetric systems, there are diamond detectors [3], ionization chambers [4-6], scintillators [7], semiconductor detectors [8], radio-chromic film [9], ESR dosimeters [10-24], thermoluminescent dosimeters (TLD) [25-28], and scintillation dosimeters [29]. Often these dosimetric systems are aided by Monte Carlo simulations that also provide many details on the particle types and energy spectra [17, 30, 31]. The recent literature includes several experiments on Fricke gels aimed at optimizing the recipe for increasing the sensitivity to dose and/or the local stability of the radiation produced ferric ions and applications for clinical beam dosimetry [32-34].

The most common matrices investigated are porcine gelatin, agarose, and sephadex [2]. Each of these systems had its advantages and limitations, but agarose was probably the most common gelling agent used for gel detectors. Furthermore, the effect of the addition of glucose, sucrose, starch, and locust bean on the response of ferrous-agarose-xylenol orange gel dosimetry has been studied [35]. Many experiments, in particular, have pursued precisely the development of a recipe for a Fricke gel that would optimize their performance as dosimeters for reading by MRI and/or optical spectrophotometers [32, 36, 37]. For this reason in some cases additive optical indicator has been added to the Fricke gel. Xylenol orange, for example, is one of the metallic ions optical indicators which is most widely used: it binds to ferric ions and gives rise to a colored complex in the visible range, whose color turns from orange to violet as the dose increases.

However, it must be considered that the diffusion of Fe^{3+} ions inside the gel matrix of Fricke gels after irradiation involves a reduction of the resolution and contrast of images (MR or optical ones) of spatial dose distribution. Among the various new matrices used to reduce this diffusion process, there is the polyvinyl alcohol (PVA) which is a polymer with a simple chemical structure, water-soluble, non-toxic and cheap [38]. PVA Fricke gels were produced by subjecting an aqueous solution of PVA to freeze-thaw cycles and these gels showed a diffusion coefficient of 0.14 $mm^{-2}s^{-1}$ [38].

Recently, a new gel formulation making use of a matrix of PVA cross-linked by adding glutaraldehyde (GTA) has shown promising dosimetric features in terms of photon sensitivity and low diffusion without the need for performing freeze-thaw cycles during the preparation process [39-41].

In this work, PVA-GTA gel samples prepared with different contents of Fe^{2+} ions were exposed to clinical 6 MV photons in the dose range between 0 and 20 Gy. Since irradiation induces variations of the spin-lattice relaxation time T_1 of the nuclear magnetic resonance (NMR) signal [34], measurements of NMR relaxometry for T_1 were performed.

It should be highlighted that one issue in the development and study of the used dosimetric gels that are analyzed through clinical MRI scanners is the fact that these scanners are usually not always available for experiments. Therefore, the possibility of performing an experimental activity for preliminary characterization in the laboratory and using clinical MRI scanners only in the last step of experimentation would favor the development of the 3-D MRI dosimetric gels. Here, we want to show the advantages of using a NMR relaxometer, which is generally used for porosymetric analysis in the field of cultural heritage diagnostics [42], to make estimates of the relaxation times of dosimetric gels. Consequently, with this instrumentation, it is possible to obtain information regarding gel sensitivity with the radiation dose.

MATERIALS AND METHODS

PVA-GTA gel

The Fricke PVA-GTA gels were prepared from a 10 % w/v aqueous solution of hydrolyzed 99% purity PVA with a molecular weight between 85000 and 124000 (Sigma-Aldrich), 1 % w/v GTA, 25 mM H_2SO_4, and 0.165 mM ferric ion indicator xylenol orange sodium salt $C_{31}H_{28}O_{13}N_2Na_4$.

For the experiments, two gels were prepared with different concentrations of ferrous ammonium sulfate hexahydrate $[Fe(NH_4)_2(SO_4)_2\ 6H_2O]$ (*i. e.* 0.5 mM and 2.5 mM) in order to evaluate the changes observed with a varying amount of Fe^{2+} ions. The PVA dissolution in water (70 % of total water) was performed in CEM STAR System 2. The system was set to boil for 40 minutes to totally dissolve the PVA. A condenser was used to prevent evaporated water losses. When the solution was retrieved from the CEM STAR System 2, the PVA solution was mechanically stirred for a few minutes until homogeneous.

The water-PVA solution was allowed to cool down to 25 °C under stirring. When the PVA solution had reached a temperature of 25 °C, the Fricke solution (20 % water, sulfuric acid, ferrous ammonium sulphate, and xylenol orange sodium salt) was added to the PVA solution. The mixing was improved by gentle stirring. When the combined solution (PVA solution with Fricke solution) had reached homogeneity, the remaining 10 % of the GTA solution was added. The GTA solution was made with a 9 % v/v of water and 1 % v/v of GTA. The final concentrations of the reagents were: 10 % w/v PVA, 1 % v/v GTA, 25 mM sulphuric acid, 0.5 mM or 2.5 mM iron ammonium sulphate, and 0.165 mM xylenol orange. The solution was kept under stirring for a few more seconds to homogenize it, then it was poured into polystyrene molds. Ultrapure water (UPW) (resis-

tivity 18.6 M cm) was used for the preparation of gels. Dosimetric gels were stored in the dark under refrigeration (10 1°C) after preparation and between irradiation and measurement, in order to minimize possible oxidation of Fe^{2+} ions induced by temperature or light.

Irradiation

Fricke gel samples were irradiated at doses between 0 Gy and 20 Gy by 6 MV X-rays generated by a Siemens Primus (Siemens Medical Systems, CA, USA) at the radiotherapy department of A.R.N.A.S. Civico, Palermo, Italy. The dosimetric gels were exposed to a 35 cm × 35 cm photon field.

A build-up layer was placed above the samples and 5 cm water equivalent layers were placed below the samples. All photon irradiations were made under electronic equilibrium conditions. The photon absolute dose values were measured using an ionization chamber (Semiflex type 31010™, PTW: active volume 0.125 cm^3, active length 0.65 cm) following the IAEA TRS-398.

Instrumentation

A single-side relaxometer mq ProFiler (Bruker Biospin, Italy), operating at about 15 MHz, was employed for NMR relaxometry measurements. All measurements were performed at room temperature, protecting each sample under measurement from light and covering it with a film in order to avoid any contamination of the gel. Dosimetric gels were subjected to saturation-recovery-spin echo sequences: 90° pulses and a successive pair of pulses 90°-180° given after a time T_S (saturation time), after the first 90° pulse (see fig. 1). An inter-pulse delay for spin-echo signals of 44 s, *i. e*., the shortest possible delay, was set. The signal recorded is the MR echo and the related integral is considered as a MR signal.

If multiple acquisitions are performed with increasing the saturation time T_S, the MR signal (proportional to the longitudinal magnetization M_Z) increases following an exponential saturation function according to

$$M_z(t) \quad M_\infty \; 1 \quad e^{\frac{t}{T_1}} \qquad (1)$$

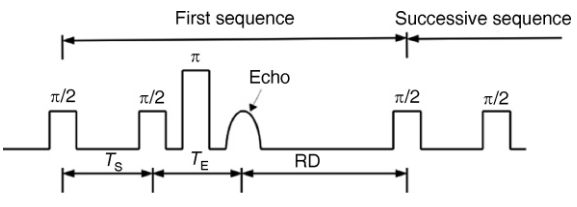

Figure 1. Schematic representation of the saturation--recovery-spin echo sequence

where M_∞ is the value of the MR signal for very large saturation times and T_1 – the spin-lattice (longitudinal) relaxation time. The value of T_1 (and that of its reciprocal R_1 named the longitudinal relaxation rate) is important from the dosimetric point of view because it is dose-dependent and is used for dose estimation.

The optimization of the NMR sequence used for the T_1 measurements was aimed at obtaining a high signal-to-noise ratio (SNR) and performing quite short readings to minimize the auto-oxidation phenomena which are favored by light exposure and other external environmental factors during the measurement process.

The parameters considered were: recycle delay (RD), the number of accumulations for a single measurement, and the total number of points with increasing the saturation time T_S.

We studied the trend of R_1 with varying the recycle delay, *i. e*. the time an after the echo and the repetition of the pulse sequence (fig. 1). The purpose of this study is to find the minimum value of RD to ensure the complete return of the magnetization to the equilibrium value and then avoid underestimation of the magnetization in successive measurements. The study was conducted by varying the RD from 0.1 s to 2.0 s with regular steps (and keeping the other parameters fixed: number of scans 16, the range of saturation time T_S from 0.1 to 10000 ms, increment factor 1.5). For this analysis, we chose to study the Fricke unirradiated gels as these samples have longer relaxation times than those irradiated and therefore are more affected by the choice of short RD. If RD value does not affect the NMR measurements in unirradiated samples then it definitely will not affect measurements on irradiated samples. The values of R_1 obtained from this series of measurements are reported in fig. 2.

As shown in fig. 2, the R_1 value is strongly influenced by the delay time. For RD values smaller than 1.0 s the measured magnetization is unable to return to the equilibrium configuration and the R_1 value extrap-

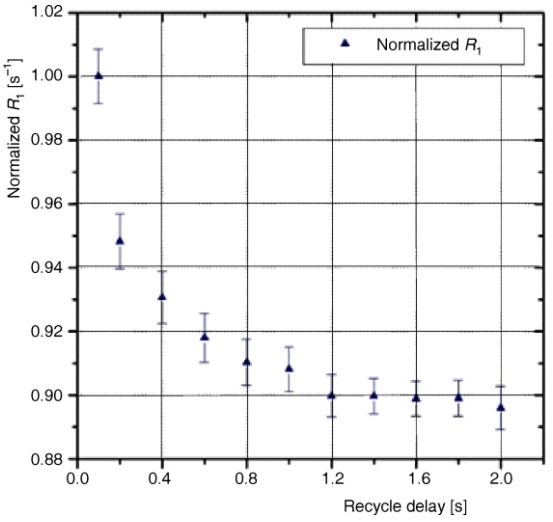

Figure 2. Trend of the longitudinal relaxation rate for an unirradiated Fricke gel with varying the recycle delay

olated from the fit is overestimated. For RD values larger than or equal to 1.0 s the extrapolated R_1 values are comparable within the experimental error.

Therefore, for further measurements on irradiated Fricke gels, a value of 1.2 s as recycle delay was chosen to guarantee the complete recovery of the magnetization with changing the dosimetric information of samples (R_1).

During the optimization phase of a NMR sequence it is fundamental to study the number of accumulations to enhance the SNR while avoiding measurements that last an excessively long time. For a fixed saturation time T_S (*i. e.* 5000 ms for which the gel NMR signal has largely reached the saturation value) we acquired individual echo signals with varying the number of accumulations to assess how the choice of this acquisition parameter influences the measurement results.

In order to assess the signal-to-noise ratio the maximum value of the echo signal was considered and the noise was calculated as the standard deviation of the MR signal where there is no echo signal (fig. 3). In fig. 4 the SNR is reported as a function of the number of accumulations.

The signal-to-noise-ratio as a function of the accumulation number was fitted using the following curve

$$SNR \quad AN^{b} \tag{2}$$

where A is a global factor, N – the number of scans and b is the exponent. The curve shown in fig. 4 was obtained by a fit and the value of the b exponent was found to be 0.45 0.04 (consistent with the expected $N^{1/2}$ dependence) with a correlation coefficient R^2 = = 0.954.

Furthermore, the complete determination of the T_1 relaxation times was performed by increasing the number of accumulations. We have found that if the number of accumulations is doubled from 16 to 32 the

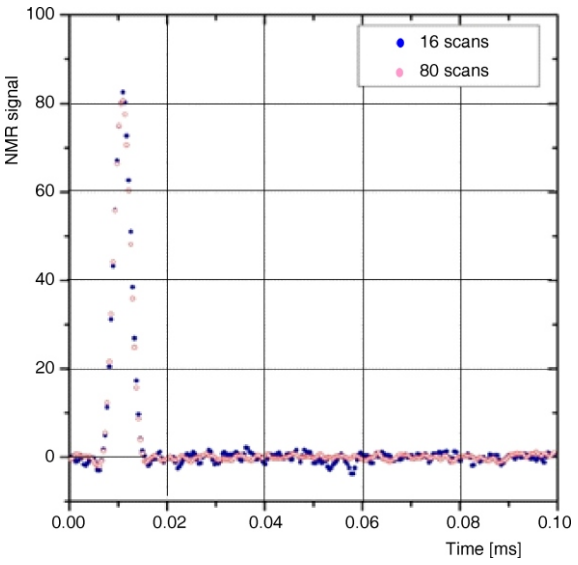

Figure 3. Experimental data of the NMR echoes recorded with increasing the number of accumulations. This is the experimental measurement of the echo reported in the scheme of fig. 1

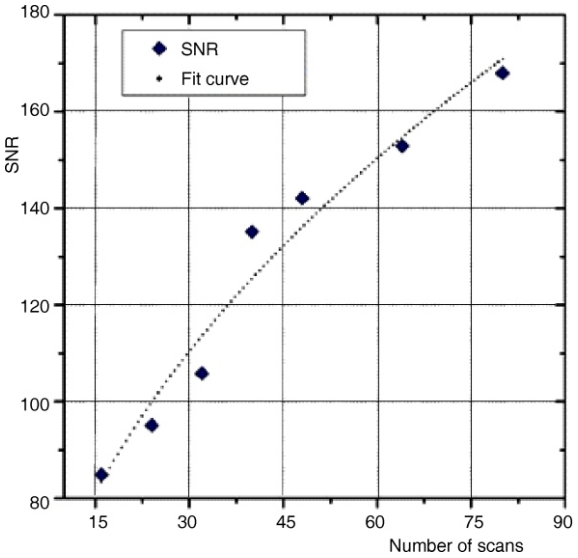

Figure 4. SNR *vs.* the number of accumulations

relative standard deviation (RSD) is reduced by 25 % and if the accumulations are increased from 16 to 64 the RSD is reduced by 35 %. A good compromise between improvement of SNR and excessive measurement time can be achieved with 32 accumulations.

In order to have a sufficient number of values (for the fitting procedure) to extract the T_1 value, we have chosen for T_S the range between 0.1 ms and 8500 ms with an increment multiplicative factor of 1.5 with 29 experimental values. The above-mentioned values allow obtaining a signal sampling with 29 values in about 25 minutes.

RESULTS AND DISCUSSION

The response of these PVA-GTA dosimetric gels with xylenol-orange for different concentrations of iron was analyzed. The experiments reported below were focused on the longitudinal relaxation time T_1 since its reciprocal value $R_1 = 1/T_1$ is dose-dependent and, in particular, linearly increases with dose [33] according to the following relation

$$R_1(D) \quad kD \quad R_1(0) \tag{3}$$

where $R_1(0)$ is the longitudinal relaxation rate of unirradiated samples (and is related to the initial concentration of Fe^{2+} ions present inside the gels) and k is a constant value characteristic of the gel dosimeter (and is related to the concentration of Fe^{3+} ions produced after irradiation of the gels).

NMR relaxometry measurements

After the choice of the best parameters for data acquisitions, the NMR relaxometer was used for dosimetric characterization of the irradiated gels. Dose sensitivity of Fricke gels prepared with 0.5 mM and 2.5 mM of Fe^{2+} was investigated. Our analysis was focused on the dose dependence of the longitudinal relaxation rate R_1.

Unirradiated samples and exposed samples of Fricke gels were studied through the saturation recovery (SR) sequence. The signals for different saturation times (T_S) describe the relaxation of the longitudinal *H*-proton magnetization M_Z from zero to the equilibrium value. Examples of these MR signals for two samples (one unirradiated and the other irradiated with photon beams at 15 Gy) are reported in fig. 5. As expected, with increasing the absorbed dose the relaxation time T_1 decreases (and consequently the R_1 increases) as shown by the faster recovery of the MR signal for irradiated samples than the unirradiated one. An exponential fit of these signals was performed using the time function

$$M_z(t) \quad G \; 1 \quad e^{\frac{t}{T_1}} \tag{4}$$

with G and T_1 fit parameters, and permitted to obtain the T_1 value for each sample.

Figure 6 compares the results of NMR relaxometry measurements on the Fricke PVA-GTA gels prepared with different concentrations of Fe^{2+} ferrous ions [34]. All data are expressed in terms of the longitudinal relaxation rate R_1 *vs*. the photon irradiation dose.

The Fe^{3+} ions produced by radiation-induced oxidation of Fe^{2+} ions act as T_1-contrast agents, making the longitudinal relaxation of *H*-proton magnetization faster.

Consequently, R_1 is an increasing function of dose. Experimental trends are well described by linear dose functions (*i. e.* $R_1(D) = a + b$ D) as confirmed by the corresponding correlation factors reported in tab. 1.

For a better comparison of the experimental parameters with varying the concentration of ferrous Fe^{2+} irons ions the slopes and intercepts are reported in fig. 7.

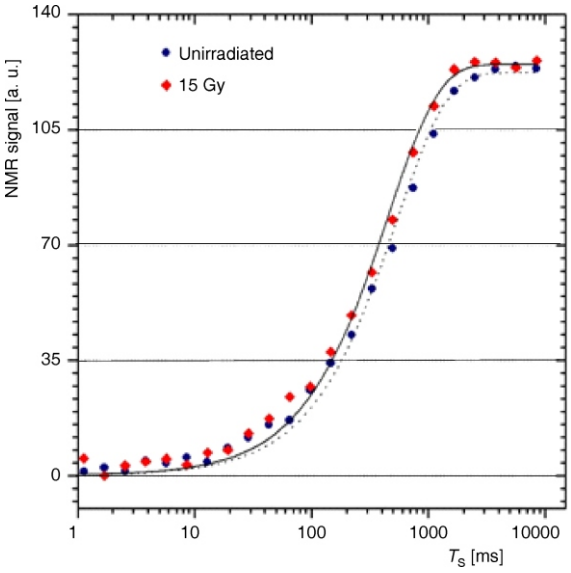

Figure 5. MR signal as a function of the saturation time T_S

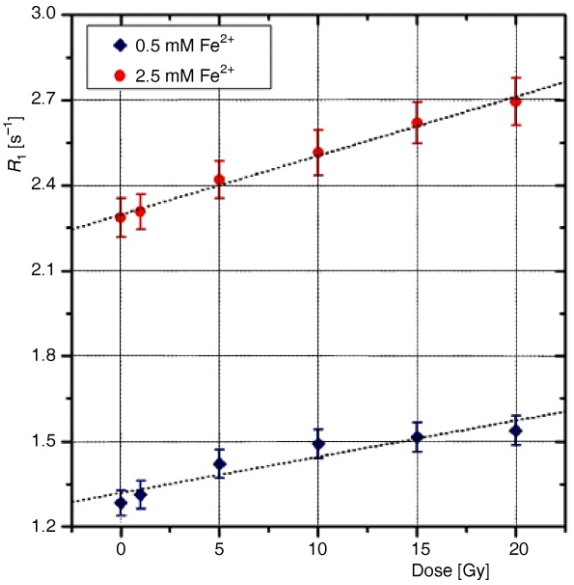

Figure 6. Longitudinal relaxation rate R_1 for PVA-GTA Fricke gels as a function of the absorbed dose

Table 1. Parameters of the linear regressions of datasets related to Fricke gel dosimeters shown in fig. 6

0.5 mM Fe^{2+}	2.5 mM Fe^{2+}
R_1 intercept = 1.31 0.02	R_1 intercept = 2.295 0.010
R_1 slope = 0.013 0.002	R_1 slope = 0.0210 0.0010
R^2 = 0.9835	R^2 = 0.9974

Figure 7 shows the intercepts and slopes as a function of the concentration of iron used. The increase of Fe^{2+} concentration in a solution increases the longitudinal relaxation rate R_1 in agreement with data from the literature [34].

This is because Fe^{2+} ions are paramagnetic and influence the hydrogen relaxation times which are measured by means of a NMR relaxometer. In particular, the increase of the relaxation rate R_1 by increasing

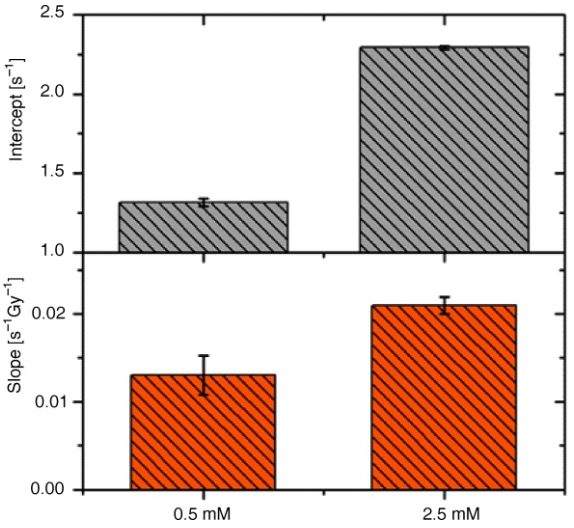

Figure 7. Slopes and intercepts of the linear regressions of datasets related to Fricke gel dosimeters shown in fig. 6.

the Fe^{2+} concentration of factor 5 is about 75 % and therefore R_1 is not directly proportional to the increase of the Fe^{2+} ions as also reported in the literature for agarose gels [34].

Figure 7 shows also the slopes of the linear trend lines reported in fig. 6 and these slopes are correlated to the sensitivity of these Fricke gels. The sensitivity increases with Fe^{2+} concentrations from 0.5 mM to 2.5 mM. The five-fold increase in the Fe^{2+} amount inside the gel matrix introduces an increase in sensitivity of almost 60 %. Therefore, in the range of the Fe^{2+} concentrations considered an increase of the amount of Fe^{2+} ions involves an increase of the dosimeter sensitivity which is always necessary for dosimetric applications.

CONCLUSIONS

In this work, we investigated the sensitivity of Fricke gels dosimeters prepared with polyvinyl alcohol added with glutaraldehyde as a function of the ferrous sulfate content.

The analysis carried out by means of NMR relaxometry pointed out that the MR signal increases as the Fe^{2+} content increases and the increase is of about 75 % when the concentration of Fe^{2+} ions is increased from 0.5 mM to 2.5 mM. Furthermore, the sensitivity improvement achieved with increasing the Fe^{2+} concentration is of about 60 %. This paper shows that a portable NMR relaxometer used for analysis of porous materials can be used for characterization of dosimetric gels and this study can be considered as the first step for the characterization of these dosimeters which in the future could be used for 3-D dose mapping in clinical applications.

ACKNOWLEDGMENTS

This research was supported by the Italian Ministry of Research and University (Project: Development and application of new materials for ionizing radiation dosimetry, P. I. F. d'Errico), by the National Nuclear Physics Institute (Project: “NextMR”, Local P. I. M. Marrale), and by the University of Palermo.

AUTHORS' CONTRIBUTIONS

The experimental work (gel preparation, NMR measurements and data analysis) was done by S. Gallo, G. Collura, A. Longo, and L. Tranchina. G. Iacoviello performed samples irradiations. The motivation behind the research was provided by both F. d'Errico and M. Marrale. The manuscript was written by M. Marrale and A. Bartolotta.

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Received on April 5, 2017
Accepted on July 10, 2017

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ПРЕЛИМИНАРНА АНАЛИЗА ФРИКОВИХ ГЕЛ ДОЗИМЕТАРА НАПРАВЉЕНИХ ОД ПОЛИВИНИЛ АЛКОХОЛА И ГЛУТАРАЛДЕХИДА, ОБАВЉЕНА РЕЛАКСОМЕТРИЈОМ МАГНЕТНЕ РЕЗОНАНЦЕ

Приказана је прелиминарна анализа Фрикових гел дозиметара које карактерише примена нове формулације матрице поливинил алкохола са додатим глутаралдехидом, извршена методом релаксометрије нуклеарне магнетне резонанце. У претходним оптичким студијама, ови гелови су испољили изгледне дозиметријске особине у погледу осетљивости на фотоне и слабу дифузију феритних јона насталих после озрачивања.

У овом раду је употребљен преносни релаксометар нуклеарне магнетне резонанце како би се измерила времена релаксација (која су важна за дозиметријске примене) гел материјала. У ту сврху, извели смо студију за оптимизацију параметара аквизиције релаксометром нуклеарне магнетне резонанце. Узорци гела изложени су клиничким фотонима од 6 MV у опсегу доза од 0 до 20 Gy. Мерења релаксација нуклеарне магнетне резонанце обављена су за различите вредности концентрација јона Fe^{2*} при чему је измерена осетљивост на сноп фотона. Анализа показује да сигнал магнетне резонанце расте са повећањем концетрације јона Fe^{2+} и да пораст износи око 75 % за повећање концетрације јона Fe^{2+} са 0.5 mM на 2.5 mM. Шта више, постигнуто је повећање осетљивости за око 60 % при повећању концентрације јона Fe^{2+}. Показано је да се преносни релаксометар нуклеарне магнетне резонанце, чија је намена анализа порозних материјала, може користити за карактеризацију ове врсте дозиметријских гелова и ова студија се може узети као први корак у карактеризацији ових дозиметара који би се у будућности могли користити за 3-D мапирање доза у клиничким применама.

Кључне речи: дозиметрија, гел, нуклеарна магнетна резонанца, поливинил алкохол, глутаралдехид, радиотерапија