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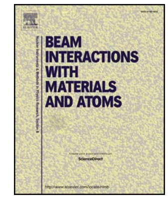

Comparative study of nuclear magnetic resonance and UV–visible spectroscopy dose-response of polymer gel based on *N*-(Isobutoxymethyl) acrylamide

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A R T I C L E I N F O

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A B S T R A C T

A comparative study of nuclear magnetic resonance and UV–visible spectroscopy of dose-response for polymer gel dosimeters was performed. Dosimeters were prepared using *N*-(Isobutoxymethyl) acrylamide (NIBMA) as a new monomer via radiation induced polymerization for use in radiotherapy planning. The prepared dosimeters were irradiated with doses up to 30 Gy at a constant dose rate of 600 MU/min. Using a medical linear accelerator at irradiation energies of 6, 10 and 18 MV photon beam. The nuclear magnetic resonance (NMR), via spin-spin relaxation rate (R_2) for water proton surrounding the polymer formulation and UV–Visible spectroscopy, via the optical absorbance measurements of irradiated dosimeters at selected wavelengths of 500 nm, was used to investigate the dose response of NIBMAGAT gel dosimeters. Scavenge of oxygen was done using tetrakis (hydroxymethyl) phosphonium chloride (THPC). The THPC optimum concentration in the dosimeters formulations were 5 and 10 mM for the NMR and optical absorbance measurements respectively. The quantitative investigation of the dosimeters components reveals the selective formulations based on 4% w/w gelatin, 1% w/w NIBMA, 3% w/w BisAAm, 5 or 10 mM THPC and 17% w/w glycerol which significantly increase the dosimeters dose response. The prepared dosimeters were found to be dose rate and photon beam irradiation energy independent. The stability study shows no change in the relaxation rate or in the optical absorbance of the gel dosimeters up to 8 days post-irradiation. The prepared polymer gel dosimeters at the energies of 6, 10 and 18 MV photon beam irradiation in the range of 1–30 Gy have the linearity of the dose response function in the case of R_2 is better than in the case of absorbance measurements; correlation coefficient (r^2) equals 0.995 and 0.991, respectively. Dose sensitivity, R_2 of NIBMAGAT dosimeters ($0.0775\ s^{-1}\ Gy^{-1}$). The absorption band intensity increases linearly with a dose sensitivity of $0.016\ cm^{-1}\ Gy^{-1}$. The detection limit of the present dosimeter analyzed by R_2 and absorbance measurements is 1 Gy and 2 Gy respectively. The overall uncertainty measurements of dose approve that by using the absorbance measurements the gel is not useful as a dosimeter like as R_2 measurements. It could be a new composition of dosimeters successfully utilized for MRI (Magnetic Resonance Imaging) for radiotherapy treatment planning.

1. Introduction

The science of dose measurement known as dosimetry, which has contributed in many irradiation processing. It is extensively used to achieve radiation dose measurements during the establishment of a new radiation process, validation of the procedure, dose mapping, and routine does process control to realize a quality control and assurance. It's essential that all measurements related to dose monitoring are detectable to national standards or international standards. Polymer gel dosimeters are a tissue equivalent material fabricated from radiation sensitive chemicals which, upon irradiation, it polymerizes in an aqueous gelatin matrix as a function of the absorbed radiation dose. Polymer gel dosimeters have been shown to be a very useful tool in the verification of radiotherapy treatment planning [1,2]. An extensive review of polymer gel dosimetry was recently presented by Baldock et al. (2010) [3]. For many decades, there have been many efforts for developing the gel dosimeters to help in the evaluation of the distribution and magnitude of absorbed dose for radiotherapy treatment planning [3–5]. Many approaches have been proposed in this attractive field of research as radiation chemistry [6], polymer chemistry [7],

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medical physics [8], dosimetry [9], computer sciences [7].

Gel dosimeters are divided into two main groups: Fricke infused gel dosimeter which is based on radiation induced oxidation of ferrous ions which modifies NMR relaxation rates and polymer gel dosimeters [10] which is based on radiation induced polymerization and crosslinking of acrylic monomers. The formation of crosslinked polymers in the irradiated regions of the gel increases the NMR relaxation rates of the neighboring water protons. The next series was the Magnetic Resonance Imaging (MRI) to measure the radiolytic oxidation in the Fricke gel dosimeters [11–13], Then a formula was developed for gelatin/acrylamide dosimeter named BANANA [14] a later modified formulation named BANG consisted of Bis, AAm, nitrogen and aqueous gelatin [15] has appeared. A fairly large number of formulations for dosimeters was developed. The most common gel matrices investigated were agarose gel [16–18], gelatin gel [19–21], PVA gel [22–24], polymer gel [25–27] many of these polymeric dosimeters had its advantages and limitations.

The aim of the current study is to introduce a new monomer N-(Isobutoxymethyl) acrylamide (NIBMA) to polymer gel dosimeters for measurements of absorbed dose of ionizing radiation named NIBMAGAT with a formulation composed of NIBMA-gelatin-THPC. This monomer was introduced by Basfar et al. as a European patent [28]. The introduced monomer contains a vinyl group capable of polymerization. A comparative study of NMR and the optical absorbance measurements of the dose response via detection of spin-spin relaxation rate R_2 for water proton surrounding the polymer formulation and measuring the absorption spectra of the irradiated gel dosimeters at selected wavelengths of 500 nm consecutively has been conducted. The performance of the dosimeters was studied at 6, 10 and 18 MV photon beam irradiation in the range of 1–30 Gy. This dosimeter is designed for radiotherapy dosimetry and falls into polymer gel dosimeters.

2. Materials and methods

2.1. Materials

All chemicals, *N*-(Isobutoxymethyl) acrylamide (NIBMA), Gelatin (Type A, bloom 300), *N,N*-methylene-bis-acrylamide (BIS), tetrakis (hydroxymethyl) phosphonium chloride (THPC) and glycerol were purchased from the SIGMA Chemical Co. (St. Louis, Mo, USA). NMR tube 1 cm diameter and 20 cm height and 3 cm disposable cuvettes were purchased from Wilmad glass, (Buena, NJ, USA).

2.1.1. Preparation of NIBMAGAT polymer gel dosimeters

The preparation of the gel dosimeters using the *N*-(Isobutoxymethyl) acrylamide polymer gels throughout this work in the atmospheric conditions without nitrogen gas was according to the methodology of Jirasek et al. [29]. The NIBMAGAT dosimeters were composed of 1–2% w/w of *N*-(Isobutoxymethyl) acrylamide (NIBMA), 1–4% w/w of *N,N*-methylene bis acrylamide (BIS), 2–5% w/w of gelatin, 2.5–20 mM THPC and 0–29% w/w of at least one co-solvent selected from the group consisting of glycerol, acetone and methanol and ultra-pure deionized water as a solvent. The quantitative description of the NIBMAGAT dosimeters composition varied according to the studied component concentrations, dose rate, radiation energy and storage time.

General procedure: before heating the gelatin to 50 °C for 1 h, it was left to soak for 10 min in the deionized water, while incessantly stirring. BisAAm, NIBMA and glycerol were added and dissolved. Addition of THPC was done when the solution reached 35 °C and stirred for 5 min. The polymer gels component was filled in NMR tubes and cuvettes, sealed and stored in a refrigerator until irradiation.

2.1.2. Irradiation of polymer gels

The cuvettes and the NMR tubes containing gels were placed in a $30 \times 30 \times 30\ cm^3$ cubic water phantom according to the IAEA protocol of external radiotherapy TRS 389 protocol with a calibrated ionization chamber [30]. Samples were irradiated with beam field size of $10 \times 10\ cm^2$ to different doses at 5 cm depth and 100 cm source-to-surface distance (SSD) using medical linear accelerator photon beam (Varian medical systems, USA) under 10 MV irradiation energy, at the dose rate 200–600 MU/min. In the current experiment, we used water phantom and a field size of 10 × 10 cm, 1 MU = 1 cGy at a depth of maximum dose, Z_max. The irradiated dosimeters in gels containing cuvettes and NMR tubes were enclosed with an aluminum foil and stored in the freezer for 24 h before NMR and spectrophotometer measurements.

2.1.3. Nuclear magnetic resonance (NMR) measurement

The NMR relaxation rate R_2 measurements were achieved using the minispec mq20 NMR analyzer, (Bruker, Germany). All polymer gel dosimeters were transferred to a temperature controlled NMR room to equilibrate to room temperature. All measurements were performed at the ambient air temperature of 22 °C. The irradiated polymer gel sample was put in the NMR tube (1 cm diameter and 20 cm height) and lowered into the magnetic box (probe head). A standard multi Spin Echo Carr Purcell Meiboom Gill (CPMG) sequence with an echo time of 1 ms and a delay of 10s between scans was used to measure relaxation time (T_2). A temperature conditioning system was used to ensure that the temperature of the dosimeters equilibrated to the scanning temperature. Three samples at each absorbed dose were measured, but no significant differences in their characteristics were found during measurements. The irradiated dosimeters in the NMR tubes were put into the magnetic box (probe head), The magnet has strength of 0.5 Tesla. The values of relaxation rate $R_2 = 1/T_2$. The temperature during measurements was 22 ± 0.5 °C [31–33].

2.1.4. UV–VIS spectrophotometer

The absorption spectra of irradiated gel samples in the wavelength range from 350 to 650 nm were measured using UV/VIS spectrophotometer, model Lambda 850, from Perkin-Elmer, USA. The response of the gel was recorded at 500 nm. Three replicate cuvettes containing irradiated gels at each absorbed dose were measured. A cuvette containing the clear gel without THPC was initially inserted in the spectrophotometer as a blank to every light absorption measurement.

3. Results and discussion

3.1. Antioxidant concentrations

Investigation of the effect of the anti-oxidant (THPC) concentrations on the NIBMAGAT dosimeters were done with the preparation of varied compositions of gel dosimeters formulations, based on 4 w/w gelatin, 1.8 w/w NIBMA, 3 w/w BisAAm, 20 w/w glycerol and 2.5–20 mM THPC, then stored in a refrigerator 10 °C. Fig. 1(A) shows the optical absorbance of polymer gels of different concentrations of THPC in the dose range 2.5–20 Gy. The error bars in all figures represent the one sigma uncertainty calculated over three different samples for each of the measurements. The same approach applies to every subsequent data presented in this work. The results of optical absorbance increase with increasing THPC concentration and the irradiation dose. The linearity was observed only for 10 and 20 mM of anti-oxidant concentrations. The selected concentration of THPC is 10 mM for the next investigation for NIBMAGAT polymer gel dosimeters using the spectrophotometer for detection. Photo 1 represents the cuvettes containing irradiated gels and the NMR tubes irradiated gels at a variable absorbed dose, the first cuvette containing the clear gel without THPC. The intensity of the white color as shown in Photo 1A is dependent on the variation of doses and dosimeters manufacturing parameters could be analyzed using a spectrophotometer or OCT (Optical Computed Tomography) [34].

Fig. 1(B) shows the dose response curves of the relaxation properties of NIBMAGAT polymer gel dosimeters for different concentrations of THPC in the dose range 2.5–20 Gy. The results show that relaxation rate