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Gamma-radiation induced synthesis of silver nanoparticles in gelatin and its application for radiotherapy dose measurements

Y.S. Soliman *

National Center for Radiation Research and Technology (NCRRT), Atomic Energy Authority (AEA), P.O. Box 8029, Nasr City, Cairo 11787, Egypt

HIGHLIGHTS

- A new radiochromic gel dosimeter based on silver nitrate is developed.
- The gel is suitable for γ-radiation treatment in the 3–100 Gy range.
- The temperature coefficient of the gel is ~0.339% per °C.
- Overall uncertainty of dose estimation is below 5% (2σ) in the 5–100 Gy range.

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ABSTRACT

A new gel dosimeter based on a radiation-sensitive silver nitrate was formulated and investigated for its potential use in γ-radiation treatment, from 3 to 100 Gy. This gel matrix is analyzed by UV–vis spectrophotometry and X-ray diffraction (XRD). Subjecting the gel to γ-rays produces Ag nanoparticles that exhibit a plasmon resonance absorption band at 450 nm. The intensity of this band increases linearly with the increase of absorbed dose up to 100 Gy. Stability of Ag nanoparticle in the dark at 6 °C is good. The overall uncertainty (2σ) of the gel dosimeter is estimated as ~4.65% in the dose range of 5–100 Gy.

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1. Introduction

Gelatin is a hydrolysis derivative of collagen and contains a number of biological functional groups like amino acids (Marois et al., 1995). Compared with collagen, gelatin has less antigenicity and more stable physicochemical properties. It is commonly used for preparation of metallic nanoparticles for some medical application which can act as a stabilizer for the nanoparticles by inhibiting coagulation and agglomeration processes (Xu and Zhou, 2008).

The metallic nanoparticles are commonly synthesized by reduction of corresponding metal ions using ionizing radiation (Li et al., 2007; Eisaa et al., 2011). Gamma irradiation of Ag^+ solutions induces the reduction of Ag^+ into metallic Ag which exhibits a visual yellow coloration and an absorption plasmon resonance band (Naghavi et al., 2010; Zhou et al., 2012; Abdel-Mohdy, 2013). The intensity of the plasmon band increases with the increase of γ-ray exposure time, which can be correlated against absorbed dose for use in radiation dosimetry.

Various gel dosimeters were previously prepared for radiotherapeutic dose application based on a radiochromic dye of leuco crystal violet (LCV) (Babic et al., 2009), Fricke–gelatin–xylenol orange (FGX) (Davies and Baldock, 2008), and some less toxic acrylate monomer (Hiroki et al., 2013). Upon γ-irradiation, the LCV dye in the gel dosimeter is converted into violet color and this color increases linearly with the increase of radiation dose to 120 Gy (Babic et al., 2009). This dosimeter showed a poor radiation dose sensitivity and high uncertainty value especially at lower absorbed doses. Additionally, its dose response function was varied significantly by irradiation temperature; the increase of response was around 13% by the increase of temperature from 15 to 20 °C (Babic et al., 2009). The uncertainty associated with dose monitoring was reported to be around 13%. Spectrophotometric measurements of FGX gel dosimeter demonstrated reproducible linear dose response only up to 25 Gy with good radiation sensitivity (Davies and Baldock, 2008). However, oxidation processes continue after γ-ray exposure influencing its dose-response function. Additionally, oxygenation during preparation enhances its radiation sensitivity but increases the natural oxidation of Fe^{2+},

* Tel.: +2 02 26714166.
E-mail address: Yasser_shabaan@hotmail.com

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diminishing the post-irradiation stability of the dosimeter. The unirradiated and irradiated FGX benzoic acid gel dosimetry showed an increase of response with the storage time by ~1% per hour during few days of storage (Bero et al., 1999). Hiroki et al. (2013) prepared different polymer gel dosimeters consisting of some methacrylate monomers (less toxic) and gellan gum. Absorbance of these dosimeters increases with increasing radiation dose by a linear function up to 10 Gy. The absorbance change in the polymer gel dosimeter is related to the degree of cloudiness (optical density) resulting from the aggregation of copolymer gel formed by radiation-induced polymerization of methacrylate monomers (Hiroki et al., 2013).

The influences of γ-rays on silver nitrate were studied widely (Naghavi et al., 2010; Zhou et al., 2012; Abdel-Mohdy, 2013), however, no attempt was initiated to investigate it for gel dosimetry in radiotherapeutic Gy levels. The main target of the present study is to introduce a new radiochromic gel dosimeter by using silver nitrate for Gy level applications and to discuss its physico-chemical properties. The Ag nanoparticles produced by irradiating the gel are characterized by UV–vis spectrophotometry and XRD spectroscopy. The influence of temperature on dosimeter performance and silver nanoparticles stability were investigated. In addition, the overall uncertainty associated with dose monitoring is estimated.

2. Experimental

2.1. Chemical

Silver nitrate (99.8%, from BDH, England), skin porcine gelatin (Aldrich) and doubly distilled water were used for preparation of silver nitrate gel dosimeter. All chemicals used in formulating the gel were of analytical grade without further purification.

2.2. Preparation of gel dosimeter

Five aqueous gelatin (8% by weight) solutions were prepared at 40 °C. Silver nitrate was added to gelatin solutions at a temperature of 32 ± 2 °C with various amounts (0, 100, 150, 200 and 250 mM) and well-homogenized. These mixtures were then pipetted into polystyrene plastic capped cuvettes ($1 \times 1 \times 3$ cm^3) and allowed to form transparent gels overnight in a refrigerator at ~6 °C. The gel cuvettes were then taken out and kept at room temperature (approximately 22 °C) in the dark until reaching thermal equilibration prior to the irradiation process.

The purpose of using gelatin is to envelop and to stabilize Ag nanoparticles formed by γ-irradiation in the silver nitrate gel. Where, gelatin can be able to interact with Ag nanoparticles by electrostatic interaction exhibiting formation of hydrogen bond (Mahmoud and Abbo, 2013). Gelatin was used with higher content % in the present study to minimize the agglomeration, mass-transfer and diffusion rate of Ag nanoparticles in the gel (Vegera and Zimon, 2006).

2.3. Irradiation and characterization of gel dosimeters

The gel dosimeters were subjected to γ-rays at room temperature using a ^{137}Cs Gamma Cell with an absorbed dose rate of 27.026 Gy/h. The absorbed dose rate of the cell was calibrated using a Fricke reference standard dosimeter (ASTM E 1026, 2004) and verified by a reference alanine dosimeter (traceable to National Institute of Standard and Technology, USA). The irradiation process was carried out in the central position of the cell that was calibrated by reference standard dosimeters. A specially designed phantom/holder of polymethylmethacrylate (PMMA) was used for irradiation of the gel cuvettes to ensure an electronic equilibrium and an adequate depth-dose build up during the process. In this process, ^{137}Cs γ-rays interact with matter in the aqueous gel mainly by photoelectric absorption and compton scattering generating free electrons and species of hydrated electrons and hydrogen radicals arising from water radiolysis (Chen et al., 2007; Naghavi et al., 2010). The electrons and hydrogen radicals reduce the Ag^+ ions into Ag^0 atoms, which then aggregated to form silver nanoparticles (Naghavi et al., 2010).

The UV–vis spectra of unirradiated and irradiated gel dosimeter were investigated using a UV–vis spectrophotometer (Evolution 500, Thermo Electron Corporation, England). In addition, the X-ray diffraction (XRD) patterns of unirradiated and irradiated gels, in a powder form, were performed at room temperature. The gels in the powder form were obtained by lyophilizing the aqueous gel samples using a freeze dryer (Lyotrap, LTE Scientific LTD in UK). The freeze drying was used to protect Ag nanoparticles produced by radiation from coagulation and to prevent reduction of Ag^+ into metallic Ag during the drying process. The measurements of powdered gel were carried out by a Shimadzu X-ray diffractometer (XRD-6000 model, 40 kV, 30 mA) equipped with an X-ray tube (Cu target). The X-ray data were recorded with a continuous scanning mode and scanning speed 8°/min in the range of 4–90° (2θ). The average particle size of Ag nanoparticles was calculated from the full width at half maximum and the peak position of XRD line broadened according to the Scherrer equation (Li et al., 2009).

$$D = 0.89\lambda/\beta \cos(\theta)$$

where D is the particle size, 0.89 is the Scherrer constant related to the shape and index (hkl) of the crystals, λ is the wavelength of the X-ray (Cu Ka, 1.54056 Å), θ is the diffraction angle, and β is the correlated full width at half maximum (FWHM in radian). The Scherrer equation is limited to the particles with nano-size scale and not applicable to grains larger than about 100 mm (Paranhos, et al., 2007).

3. Results and discussion

3.1. Absorption spectra

Fig. 1 shows the optical absorption spectra of pure gelatin samples (control) and silver nitrate gel dosimeters (200 mM) in the spectrum range of 320–750 nm for the 0–100 Gy dose range. Clearly, the control sample is colorless and has no absorption peak in this spectrum range. However, the unirradiated $AgNO_3$ gel has a pale yellow color (background) indicating the formation of Ag nanoparticles by the action of heat during preparation of the gel (Darroudi et al., 2011). Upon irradiating the $AgNO_3$ gel, its background is converted into a visual yellow color and finally to yellowish-brown as the dose increases. This color is characterized by a significant main band located at ~450 nm that corresponds to the surface plasmon resonance absorption of metallic Ag (Ramnani et al., 2007; Oldenburg, 2013). The intensity of the band increases progressively with the increase of radiation dose without appreciable shifts in the band position. Meanwhile, the peak broadening decreases gradually with increasing absorbed dose and a secondary small peak centered at ~510 nm is developed at the higher doses. The intensity of 510 nm band increases also with the increase of absorbed dose. The increase in intensity of 450 and 510 nm with the radiation dose is related mainly to the increase of Ag nanoparticles concentration in the gel upon irradiation (Zhou et al., 2012; Abdel-Mohdy, 2013). In addition, the position of the plasmon resonance band is mainly dependent on the nanoparticle size (Ramnani et al., 2007; Oldenburg, 2013) and on the surrounding medium (Yiwei et al., 2007; Xu and Zhou, 2008).

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