

Investigation upon recycling of high energy electron beam irradiated alanine dosimeter

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Abstract

This paper presents the recycling possibility of irradiated alanine dosimeter via annealing methods. The alanine samples were irradiated under 10 MeV electron beam with the doses of 10, 30, and 60 kGy. The samples were stored in an oven with the different temperature and time. The fading effects of the EPR signal of the alanine samples were investigated, and the optimum time and temperature were found. Furthermore, the calibration curves have been drawn for the annealed samples. The stability of annealed alanine has also been examined. The results show that the optimum annealing time and temperature are 30 min and 200 °C, respectively. On the other hand, it was obvious that the obtained results were independent of the radiation dose.

Keywords: EPR dosimetry, alanine, recycling, annealing.

Introduction

Nowadays, the radiation processing of materials and commercial products to enhance their quality level has become a profitable business. The most important effects of radiation on materials are the change of some properties of polymers, modification of semi-conductor components, sterilization of medical devices, destruction of parasites, insects and disease factors in foods to increase their shelf life. Radiation processing of materials is mainly performed by gamma rays and high energy electron beam. What is important in providing the desired result is the appropriate and sufficient amount of energy received by the unit mass of the material which is called the radiation absorbed dose. Different dosimetry methods are used to determine the amount of the absorbed dose. Dosimeters are usually put between the under irradiation product and the dose value received by the substance or product which can be determined. One of the very accurate and standard dosimeters is alanine. L- α -alanine is an amino-acid with a chemical formula of C₃H₇NO₂. Alanine has been the most common substance used for EPR dosimetry in the last 20 years (Regulla, 2000). Since 1962, alanine has been used in radiation dosimetry (Phil *et al.*, 2001). The first accurate dosimetric results using alanine in high-energy photons can be traced back to the activities of GSF Research Institute in Munich, Germany, between 1973 and 1974. During the years of 1952 and 1971 scientists had done research on solid organic compounds for radiation detection (Regulla *et al.*, 1984; Bergstrand, *et al.*, 1998). The EPR signal from the amino-acid L-a-alanine exposed to ionizing radiation shows an applicable relationship with the absorbed dose, and the radicals fade very slowly (Regulla *et al.*, 1982; Sleptchonok *et al.*, 2000; Dolo *et al.*, 2005). Generally, this technique represents the best method to achieve an accurate proportionality between the concentration of free radicals induced in the material due to the radiation, which was characterized by the amplitude, and the radiation dose. Considering the high price of this material, in the present work the possibility of the irradiated alanine

recycling was investigated. Thus, according to the fact that the greatest need in commercial irradiation of products is the dose range of 10-60 kGy, special attention was devoted to this interval.

Experimental procedures

Sample preparation and irradiation

The L- α -alanine powder made by Sigma Co., Germany, containing a mixture of different crystal size was used in this experiment. The powdery alanine was packed in a plastic package and the air inside was evacuated. The packages placed in a polystyrene phantom were irradiated along with a polystyrene calorimeter to measure an accurate absorbed dose. Irradiation was performed within the dose range from 10 to 60 kGy at the dose rates of about 630 kGy/min, using the 10 MeV electron beam of a Rhodotron TT200 type electron accelerator, IBA, Belgium. The accelerator was provided with a variable-speed conveyor, to pass the materials through the swept beam.

Annealing procedure

Annealing was performed using an EHRET oven Model TK/L4250, with a maximum temperature of 300°C±1. In order to avoid exposure to air during the heating, the samples were packed in an aluminum foil. The samples were annealed at different temperatures from 150°C to 220°C, in different times of 10-120 min. Annealing operation was undertaken after reaching the desired temperature and stability.

EPR measurement

The irradiated samples were carefully weighed with 0.0001 g of precision and were poured into quartz thin wall EPR tubes (4 mm in diameter) and measured with a Bruker EMS-104 spectrometer operating in X-band. The EPR spectrometer parameters used for this study are depicted in Table 1. The EPR signal intensities were measured as peak-to-peak height for the most intense

EPR lines (first derivative of the absorption spectra) per sample mass.

Table 1. The EPR parameters used to set up the spectrometer

Power	1.25 mW
Sweep Width	100 G
Modulation amplitude	2.01 G
Sweep Time	10.49 S
Filter T.C	20.48 mS
Receiver Gain	24 dB
Number of Sweeps	4
Sample Height	16 mm

FTIR spectroscopy

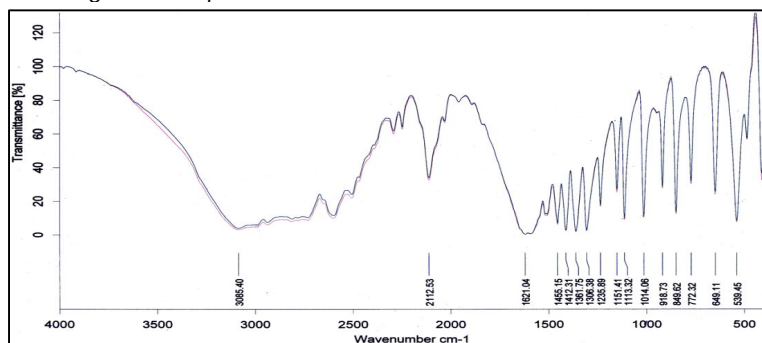
Fourier transmission infrared spectroscopy (FTIR) spectra, was carried out on irradiated samples within the wave number range of 400-4000 cm^{-1} using a Bruker EQUINOX 55 spectrometer.

Results and discussion

Characterization of irradiated alanine

The radiation induced molecular scission rate which occurs in the alanine dosimeter was investigated using FTIR spectroscopy system. Generally, the use of FTIR spectroscopy requires the preparation of a pill containing a mixture of the main sample and KBr powder as a holder material which is transparent in the range of the FTIR spectrum. Considering the fact that the sample absorption in FTIR spectroscopy is dependent on the three factors of concentration of materials, thickness and type of materials constituted in the made pill, the FTIR spectroscopy is a qualitative analysis. Therefore, to compare the irradiated and non-irradiated alanine, it is necessary that the effect of all the mentioned factors affecting the FTIR absorption should be eliminated. For this purpose, the KBr pills containing alanine were prepared and subjected to FTIR analysis beforehand, and the results were compared with the one irradiated at 60 kGy dose. Thus, in this way the effect of radiation could be studied individually. The effect of radiation on the pure KBr was also evaluated in 60 kGy of dose. The FTIR spectrum of irradiated pure KBr did not show any significant change. By comparing the FTIR spectrum of the irradiated with non-irradiated alanine no significant

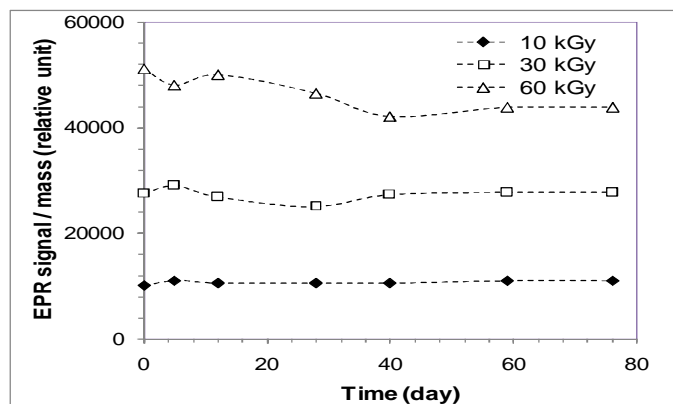
Fig. 1. FTIR spectra of irradiated and non-irradiated alanine



changes in location and intensity of the peaks was observed (Fig.1). The FTIR spectrum of the samples annealed at 200 °C was also checked which revealed that the differences were negligible.

Since the broken bonds and creating radicals are clearly confirmed by the EPR system, the lack of observed differences between FTIR spectra obtained should be somehow justified. Basically, the peak intensities in the FTIR spectra are related to the existence of bonds in the material. If the number of broken bonds after irradiation would be considerably lower than the normal bonds, any significant difference between the FTIR spectra of the irradiated and non-irradiated alanine could not be observed. In order to check the accuracy of this theory, the ratio of the broken bonds due to the radiation to the initial one at a certain amount of the absorbed dose was calculated. Considering the 2 mg of alanine and its molecular mass of 89.1gr/mol, the number of alanine molecules will be 1.35×10^{19} . The 60 kGy absorbed dose in alanine is equivalent to $6 \times 10^4 \text{ J/kg}$, therefore the energy intake by alanine molecules and consequently by each alanine molecule will be 0.12 J and $7.5 \times 10^{17} \text{ eV}$, respectively. If the energy required for breaking a bond in alanine molecule is considered to be about 20 eV, the number of alanine molecules broken is estimated to be 0.28% out of the entire molecules. The

Fig. 2. EPR signal intensity variation of the alanine sample over the time



calculation indicates that the ratio of radicals to total molecules is very low and is out of quantitative accuracy range of FTIR system.

Stability of free radicals in irradiated alanine samples

The 10 MeV electron beam irradiated samples were kept in a place far away from light and moisture at room temperature and were objected to EPR measurement in different time intervals. Fig.2 shows the stability of free radicals induced to the alanine samples irradiated in doses of 10, 30 and 60 kGy up to 2.5 months after irradiation. The results are in good agreement with those obtained in other

literatures using the x-ray and gamma-ray radiations (Phil *et al.*, 2001; Maltar-Strmecki *et al.*, 2005) This confirms alanine as a proper dosimeter for electron beam dosimetry.

Effect of annealing time and temperature on EPR responses of the irradiated samples

Electron beam irradiated alanine samples in different doses were packed into the thin aluminum foil to avoid the direct exposure to air. The oven was set on the desired temperature and the samples were placed inside the oven after stabilization of temperature. Then the samples were cooled at room temperature and their EPR signal intensities were measured and investigated. Fig.3 shows the EPR signal intensity of the irradiated alanine in different doses as a function of annealing temperature at constant annealing time of 30 min. As it can be observed, the fading effect is minimal below 150°C and becomes appreciable afterward. Since the starting melting point of alanine is 220°C, the experiment was continued up to 210°C but the results did not show any particular change within the range of 200°C to 210°C. Therefore, the optimum annealing temperature for recovery of alanine was determined in this area. This temperature is also independent of the radiation dose in the tested range of the present work.

In order to obtain the optimum annealing temperature, more than 15 different temperatures in the range specified earlier with the annealing period of 10 to 60 minutes were tested. The EPR signal intensity via annealing times for different annealing temperatures is demonstrated in Fig.4. These figures indicate that the temperature of 190°C does not seem appropriate to reduce the EPR signal intensity to the levels of non-irradiated sample. At temperature of 200°C and higher with respect to at least 30 minutes annealing time, the desired result is achieved. Therefore, the optimum annealing temperature of 200°C and annealing time of 30 min are reported as the optimum values. The results also show that the optimum temperature and time does not depend on the radiation dose. On the other hand, the

Fig.3. EPR signal intensity variations via annealing temperature for three different doses (constant annealing time of 30 min)

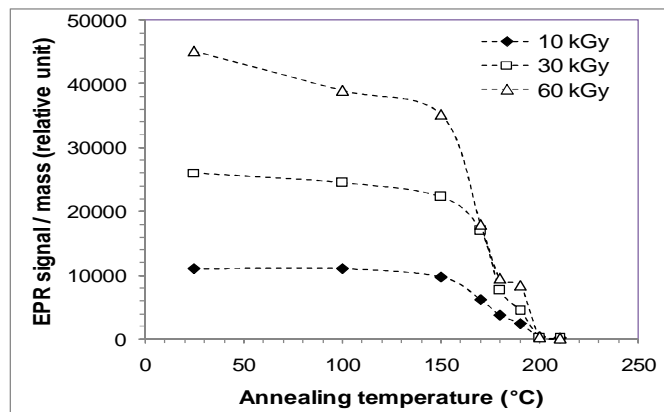
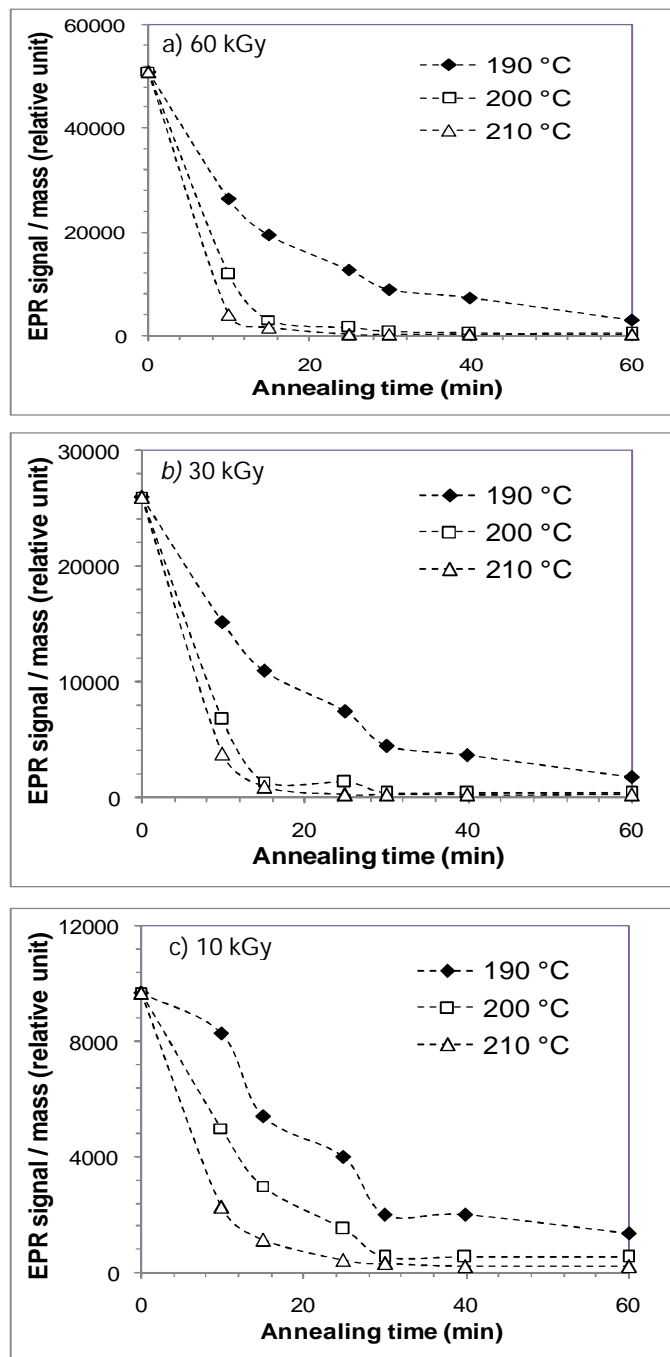


Fig.4. EPR signal intensity of the alanine samples via annealing time in different doses of, a) 60 kGy, b) 30 kGy, c) 10 kGy



reduction rates of EPR signal intensity is considerably higher in further doses. The reason for this is the exponential of radical recombination function. Thus, it can be concluded that the optimum annealing values will be more effective in higher doses.

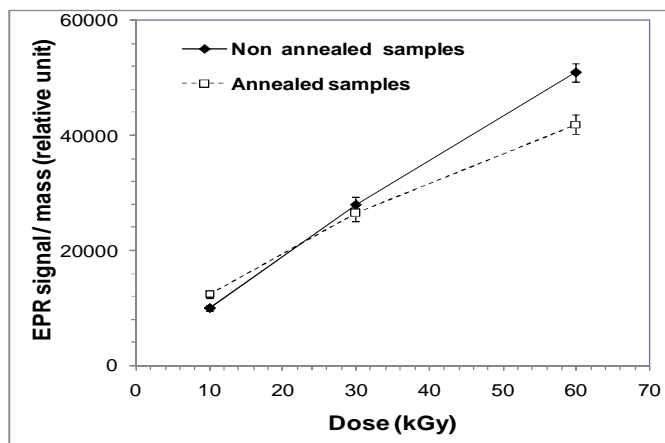
EPR signal intensity dependence on radiation dose

In this stage the irradiated alanine samples were annealed in optimum annealing temperature and time



and reused as a dosimeter. The variation of the EPR signal intensities as the function of radiation absorbed dose are shown in Fig.5 for non-annealed and annealed alanine samples. The obtained results as demonstrated in Fig.5 show that the calibration curve of the annealed alanine is almost linear in dose range of 10-60 kGy, and it can be reused as an appropriate dosimeter.

Fig.5. EPR signal intensity variation via the radiation dose for the non-annealed and annealed alanine (3 times) samples



Conclusion

Alanine dosimeter is recyclable through annealing method and its calibration curve up to three times of annealing remains linear in the range of 10-60 kGy. According to the obtained experimental results, the optimum annealing temperature of 200°C in annealing time of 30 min would be appropriate to recycle the alanine dosimeter. This temperature does not depend on the radiation dose value in the tested dose range region. The number of free radicals created within the entire alanine sample was negligible in comparison to the number of alanine molecules in the irradiated sample hence the FTIR spectra of the irradiated and non-irradiated alanine were not recognizable.

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