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Effect of Gamma-Irradiation on the EPR Study of Epoxy-Resin Films: Potential Use in Dosimetric Application

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Abstract: ***The present paper reports the results of EPR technique of gamma-irradiated epoxy-resin films. In this work, the samples have been the subject of low doses varying from 100 Gy to 1000 Gy as well as high doses ranging from 1 to 60 kGy. EPR signal spectra of both non-irradiated and irradiated epoxy resin are attributed to the superposition of strong singlet and broad spectrum comprising of the wings triplets of almost equal line intensities. The latest may be assigned to the interaction between unpaired spin and nitrogen atom. A linear behavior between the absorbed and the intensities of the EPR signal was observed for low doses, but the time stability of the epoxy resin samples was investigated for 80 days at room temperature. For the entire dose range, the signal loss did not exceed 1%, this percentage is required for routine dosimeter especially for 100 Gy- 60 kGy irradiation domain.***

Keywords: epoxy resin; gamma radiation; EPR.

1. Introduction

Epoxy resins are widely used in many industrial applications [1-3], because of their good thermal and optical properties such as in encapsulation of electronic circuits. In certain applications this organic material can be exposed to ionizing radiation which affects its electrical and optical properties [4-6]. This radiation induces indeed a modification of the physic-chemical properties especially the optical, electrical and also magnetic properties. Particular interest has been focused on the optical properties and paramagnetic due to the application of these polymers in the irradiation process. The radiation effects on the paramagnetic properties result in a change in EPR signal due to the formation of free radicals.

The development and characterization of a reliable and inexpensive precise dosimeter available in large quantities for the process control in industrial radiation processing presents an effective alternative to current dosimeters existing in the marketplace. To extend the application of epoxy-resin in the process control in industrial radiation, we investigate herewith some physical properties of such resin under irradiation.

The modifications induced by radiation in epoxy-resin have been studied extensively by many authors [7-9]. However, there is few attempts on the effect of irradiation on changes in Paramagnetic properties of epoxy-resin.

This work deals with some experiments on the paramagnetic properties of epox resin. These properties have been studied in terms of both low and high gamma irradiation doses for possibly application as routine dosimeter or as an indicator of irradiation. The signal of epoxy resin was measured with an EPR spectrometry before and after irradiation, and concentration of free radicals was calculated as function of the irradiation dose.

2. Experimental

2.1. Sample preparation

The investigated epoxy polymer was prepared by mixing an epoxy resin and hardener. The epoxy resin correspond a 2,2-bis (4 glycidyloxyphenyl) propane (molar mass 340g/mol) $C_{21}H_{24}O_4$, called epoxy resin, whose molecular structure is shown in fig.1, added with difunctional amine, supplied by Maestria, as a hardener.

After the mixture of the base resin and hardner (100:39), the compounds were carefully and homogeneously mixed. The viscous liquid obtained was poured into cylindrical silicone mould. The polymerization of the sample was carried out by a heat treatment until 150°C. The cylindrical samples obtained have 14 mm in diameter and 1 mm in thickness. They are sealed in polyster/aluminium foil/polyster/polythene sachets to avoid the effect of humidity and the absorption of the light.

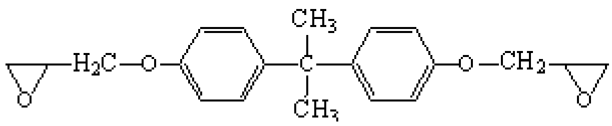

Figure 1: Molecular structure of epoxy-resin

2.2. Irradiation facility

The irradiation was carried out using the Tunisian Cobalt-60 irradiation facility at the dose rate of 115 Gy/min. The traceability to Aerial, the Secondary Standard Dosimetry Laboratory, was established using the Alanine/ESR dosimetry system. Alanine dosimeters are irradiated by the facility and returned to Aerial for dose rate assessment. The standard Fricke dosimeter was used to check the dose rate before the EPR study experiments.

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2.3. Instrumentation and procedures

EPR measurements were made by means of Bruker EMX spectrometer X-band. The measurements were recorded at room temperature. The operating conditions are: microwave power = 0.632mW, modulation amplitude =0.4 mT, modulation frequency = 100 kHz microwave frequency = 9.774 GHz and time constant = 81.920 ms .The influence of stability of EPR spectrometer setting parameters varies with time, for this raison decay was checked using a reference alanine dosimeter [10,11]

3. Results and Discussions

3.1. Radiation low dose response

Fig.2. shows the ESR spectra of epoxy resin before and after irradiation with low doses between 100 and 1000Gy. These spectra contain a single peak have a g-factor around 2.0055 centred between two triplets assigned to interaction of spin singlet and radicals formed from nitrogen hardener [12, 13] atoms. The central peak even in non- irradiated samples revealed the presence of radicals that are activated when the irradiation occurs. These radicals are phenoxyl groups (phenyl, ethyl diphenyl , phenoxy , diphenyl methyl) which their molecular structure are given in fig .1.

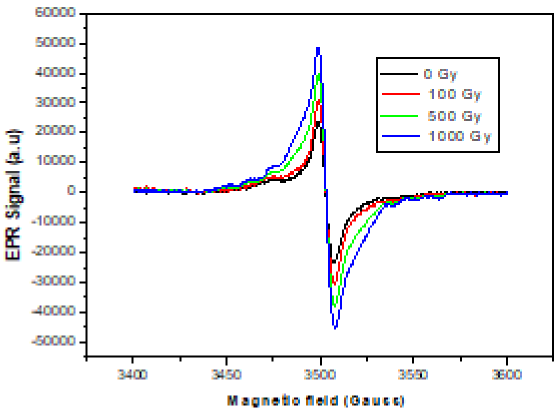

Figure 2: Amplitude of the EPR Signal as a function of low doses

A plot of signal EPR intensity as a function of gamma dose is shown in Fig. 3. From these experiments, it is noted that the signal response varies linearly with the dose under a relatively high correlation factor (R-square equal to 0.99). This linear behavior is required for probably use of such resin as a powerful dosimeter especially for low gamma irradiation doses.

The minimum sensibility for this resin is about 100Gy. This dose is so interesting for carthography experiments in order to reach the distribution manner in products subjected to gamma irradiation.

Nevertheless, EPR technique has been also tested regarding high exposition doses to explore the sensitive domain of such resin.

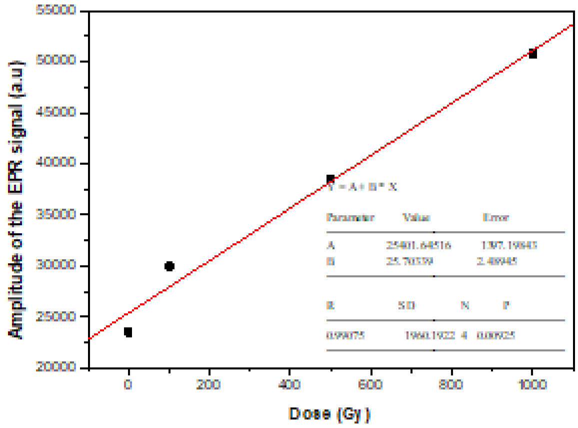

Figure 3: The EPR signal curve according to the dose

3.2. Radiation high dose response

Fig.4. shows the EPR spectra of epoxy resin before and after irradiation with high doses using large irradiation domain lying from 1 to 60 kGy. These spectra contain a single peak having a g-factor around 2.0055 located between two triplets assigned to interaction of spin singlet and radicals formed from nitrogen hardener atoms [12, 13]. Moreover, the central peak is attributed to the superposition of phenoxy radicals such as: ethyl diphenyl , phenoxy , diphenyl methyl. Their intensities are so visible when the dose is increased.

This increase of the signal response follows a polynomial law of degree 2 in terms of gamma irradiation dose, with a correlation factor (R-square of the order of 0.99). In the same line, a saturation phenomena is occurred at 50kGy. In comparison with other spectroscopic analysis techniques related to photoluminescence and colorimetric investigations, the maximum doses detected were 40 and 20 kGy respectively [14, 15].

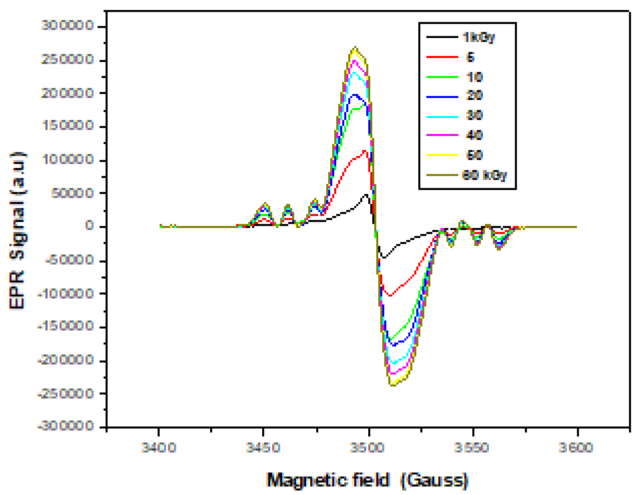

Figure 2: Amplitude of the EPR Signal as a function of high doses

Epoxy resin films have clear change in the dose range 100 Gy -60 kGy reflecting their suitability for possibly use as irradiation indicator agent. The good linearity for the dose dependence up to 1 kGy reveals this propriety. Hence, the development of a new dosimeter for quality assurance in some radiation facilities can be performed using Tunisian Co-60 irradiation facility [16].

3.3. Post – irradiation stability for high and low doses

Different factors which affect accurate routine dosimetry in radiation processing are given in [16-18]. Such parameters include irradiation temperature, humidity, radiation energy, radiation type, light, dose rate and post-irradiation stability. Among these factors, we focused our attention on the stability of EPR signal with time. Epoxy resin films packed in hermetic sachet were irradiated to 500 Gy, 1000 Gy, 10, 20 kGy. They were stored at 40% RH and room temperature (15-25 °C). EPR signal amplitude was followed during 60 days after irradiation. It is clear in fig.5 , that the normalized responses for both 1000 and 500 Gy doses of the films increase gradually that tends to be stable to the end of the storage time. The relative changes in responses did not exceed 9% after 60 days. The coefficient of variation per day is 0.15%, remains largely less than 1% (value accepted for routine dosimetry). It should also be noted that a stability after 5 days due to loss of unstable radicals is reached. This stability can be accelerated by adjusting the kinetic thermal effect, for 10 and 20 kGy γ-doses, the normalized responses of the films shows an overall decrease of EPR signal of about 35% and 55% for 10 and 20 kGy respectively. The loss per day is estimated to be of the order of 0.69% lower than 1% , value close to classical response related to usual dosimeter available actually.

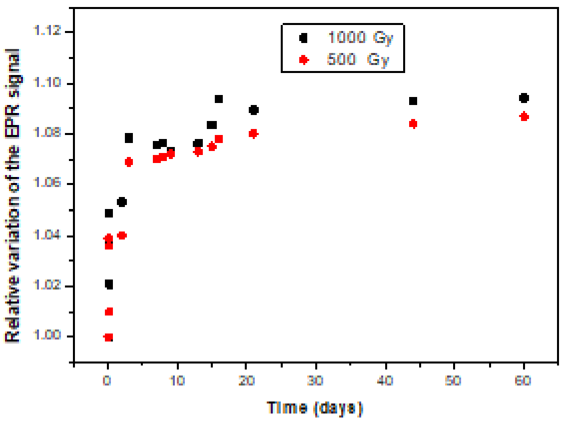

Figure 5: Kinetics of decay 500 Gy and 1000 Gy

4. Conclusion

This work reports the effect of gamma-irradiation on epoxy-resin films for low and high doses. It is found that a significant–dependent change in paramagnetic properties have been occured. These changes were analyzed by EPR measurement and the sensitivity of the dosimetric property was determined. We also demonstrate that EPR measurement is very sensitive to the variation of the signal and can be used as a method not widely recognized as a method to estimate the radiation dose. For use as a dosimeter other influencing parameters should be performed (humidity, dose rate, energy radiation and irradiation temperature). Further studies are in progress to minimize these losses as a function of the temperature. This study is so interesting since an epoxy resin is tested as a possibly powerful apparatus for gamma irradiation dosimeter.

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