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Study of L-Glutamic Acid in Solid State for its Possible Use as a Gamma Dosimeter at Different Temperatures (77, 195 and 295 K)

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Abstract The experimental response of the dosimeter as a function of the irradiation temperature plays an important role, and this effect has consequences in the practical applications of dosimetry. In this work, L-glutamic acid (2-aminopentanedioic acid) is proposed to be a good response, easy to handle, and a cheap gamma dosimeter. For this purpose, polycrystalline samples were irradiated with gamma rays at 77, 195, and 295 K and doses in the kiloGray range (43–230 kGy). The potential use of the glutamic acid system as a chemical dosimeter is based on the formation of stable free radicals when the amino acid is exposed to ionizing radiation. The observed species in these experiments were attributed to deamination and decarboxylation reactions that were studied using electron spin resonance (ESR). The results indicate that the analysis generates a linear response as the irradiation dose increases in a reliable range for industrial and research purposes at three different temperatures.

Keywords: Dosimeter, gamma radiation, L-glutamic acid, ESR

1. INTRODUCTION

Gamma radiation originates from the disintegration of radioactive nuclei, is extremely penetrating, and has been used as (1) a simple, safe, and highly effective sterilization method for biological tissues [8]; (2) as a method in food preservation [4]; and (3) in the treatment of plastics and other products [1]; also, some other applications involve irradiation at low temperatures, and

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examples of this include the irradiation of some tropical fruits and the graft copolymerization of various chemical compounds in polyethylene fibers [3]. Radiation at low temperatures prevents the radicals produced by irradiation from reacting with one another and thus increases the efficiency of other reactions [7].

L-glutamic acid (LGA) is an amino acid used by almost all living beings as a potential source of energy, and it is important in brain metabolisms. The radiation chemistry of amino acids has been studied because of the biological significance of these molecules, and these studies have been applied in different contexts, for example, in gamma dosimetry [6] or chemical evolution [5]. When organic matter is exposed to gamma radiation produce free radicals, which can be identified and quantified through electron spin resonance (ESR) analysis. The spectra can provide information about the distribution of spin density through the hyperfine structure [9].

The purpose of the present study is to examine the behavior of LCA as a possible dosimeter over the dose interval from 43 to 230 kGy at 77, 195, and 295 K using the ESR technique.

2. MATERIALS AND METHODS

2.1 Glassware and chemicals

The glass materials were treated with a warm mixture of HNO_3 and H_2SO_4 for 30 minutes, and then they were washed with distilled water and heated in an oven at 300 °C overnight [2]. LGA of the highest purity available was purchased from Sigma-Aldrich Co, USA, and was used without further purification.

2.2 Preparation of samples

2 g of LGA were placed in a glass tube inside a Dewar flask at different temperatures in the presence of oxygen.

2.3 Irradiation procedure

A Dewar flask containing the glass tube with a powder sample of amino acids was exposed to gamma-irradiation from a ^{60}Co gamma-ray source (Gammabeam 651 PT facility at the Instituto de Ciencias Nucleares, UNAM). The absorbed doses were between 43 and 230 kGy at a fixed position with a dose rate of 247 Gy/min. The Dewar flask was irradiated at room temperature (295 K), dry ice temperature (195 K), and liquid nitrogen temperature (77 K).

2.4 Analysis

200 mg of the sample were analyzed using electron spin resonance (ESR) after irradiation in a quartz tube at room temperature. The ESR spectroscopy was carried out in a JEOL JES-TE300 spectrometer at the Instituto de Química, UNAM. We used a operating in the X-Band at 100-kHz modulation frequency with a cylindrical cavity in mode TE_{011} that was equipped with a variable temperature unit. The external calibration of the magnetic field was made using a precision gaussmeter, JEOL ES-FC5. The spectrometer settings for all spectra were as follows: microwave power 1 mW; center field 335±10 mT; microwave frequency 9.44 GHz; modulation width 0.025 mT; time constant 0.1 s; amplitude 250; sweep time 120 s; and 1 scan. The readings were taken at a vertical peak height. The analyses were performed at room temperature.

The irradiated LGA samples were also analyzed at room temperature in a Perkin Elmer ATR-FTIR Spectrophotometer Spectrum 100.

3. RESULTS AND DISCUSSION

3.1 ESR data

The resulting ESR signal for LGA consists of several lines caused by the formation of various free radicals (Figure 1). They appear in the dose

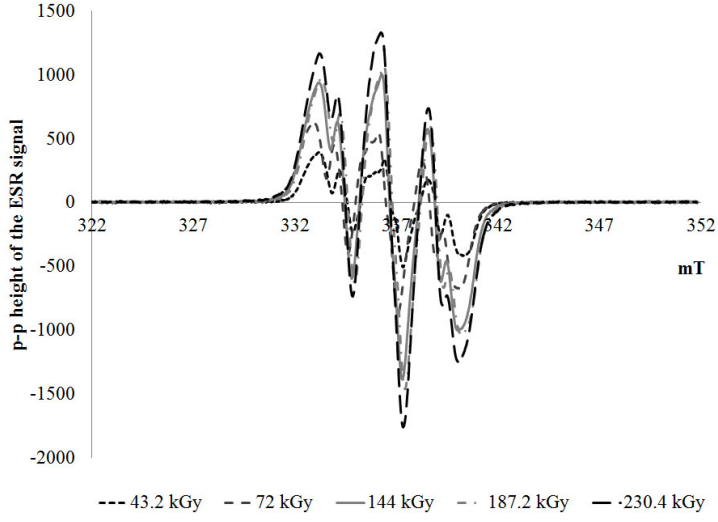

Figure 1: ESR spectra of samples irradiated at 295 K with different doses (43.2 to 230.4 kGy).

interval 43 and 230 kGy at 77, 195 y 295 K. Some of them are stable at room temperature and can be used for dosimetry purposes. The possible route to get the free radicals was a deamination and decarboxilic reaction via free radicals. The ESR signals could be assigned to the LGA decarboxilated radical from the alpha carbon of glutamic acid (HO_2C-CH_2-CH_2-CH–NH_2) and another deaminated radical (HO_2C-$\dot{C}H$-CH_2-CH_2-CO_2H). The suggested sequence of radiation-induced processes in LGA is presented in Figure 2.

3.2 Effect of the irradiation temperature

The irradiation temperature affects the ESR response. When the temperature is raised from 77 to 195 and then to 295 K, the population of radicals increases. However, the same species were observed (Figure 3). Figure 4 shows the dose-response behavior, measured as the peak-to-peak height vs. irradiation dose. Curves for 295, 195, and 77 K are represented as linear curves as a function of the dose with correlation coefficients greater than 0.9. The saturation of the species was not observed even 230 kGy.

The ATR-FTIR (Figure 5) analysis shows four principal bands. The first one is at 3012 cm^{-1}, corresponding to the vibration of the O-H of carboxylic acid (1). The second one is the vibration of the C=O present in the carboxylic acid (2), which appears at 1637 cm^{-1}. The third one is at 1504 cm^{-1}, corresponding

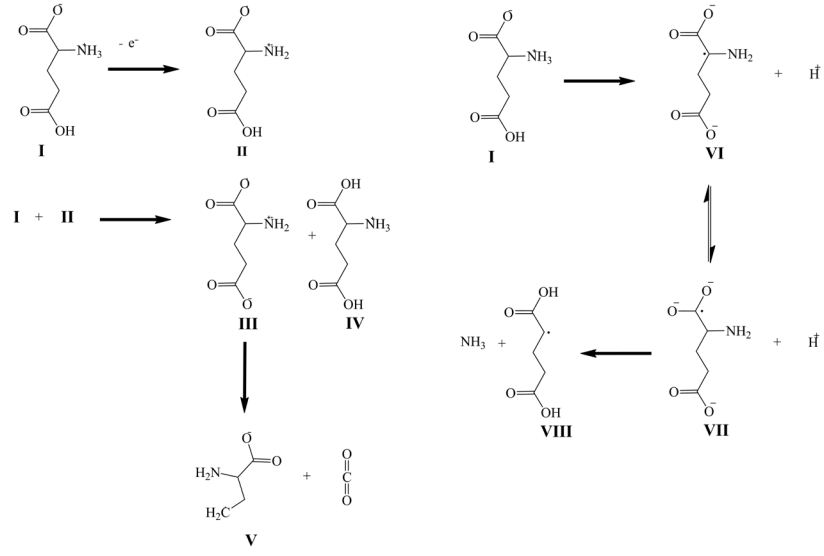

Figure 2: Suggested sequence of radiation processes for the formation of radicals V and VIII.

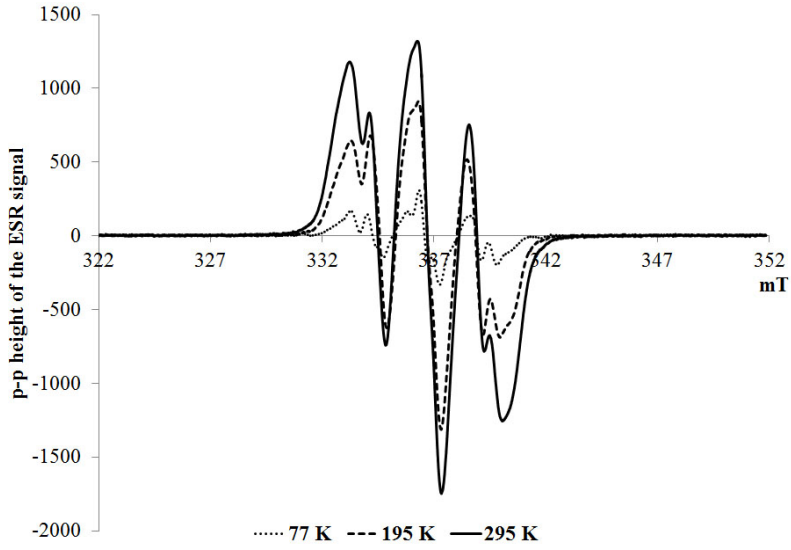

Figure 3: ESR spectra for samples irradiated at 77, 195, and 295 K at 230 kGy.

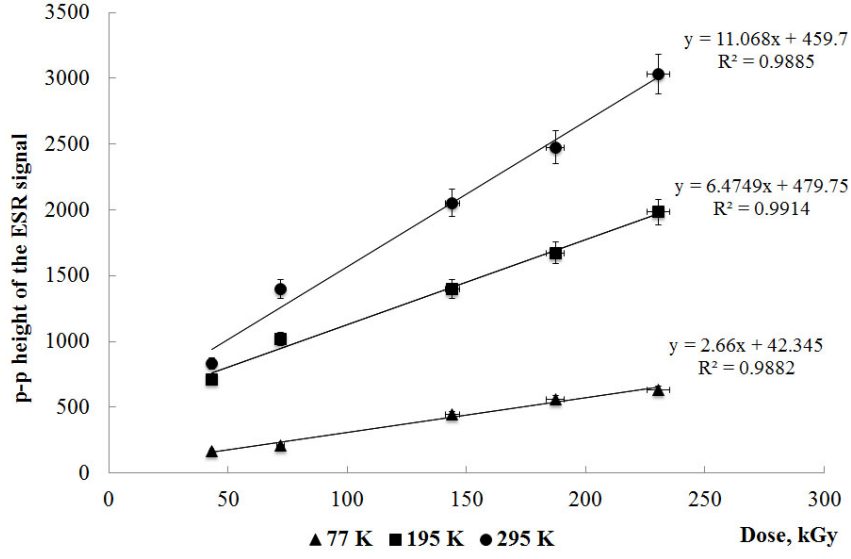

Figure 4: Linear range interval of the dose–response curves irradiated at different temperatures and doses.

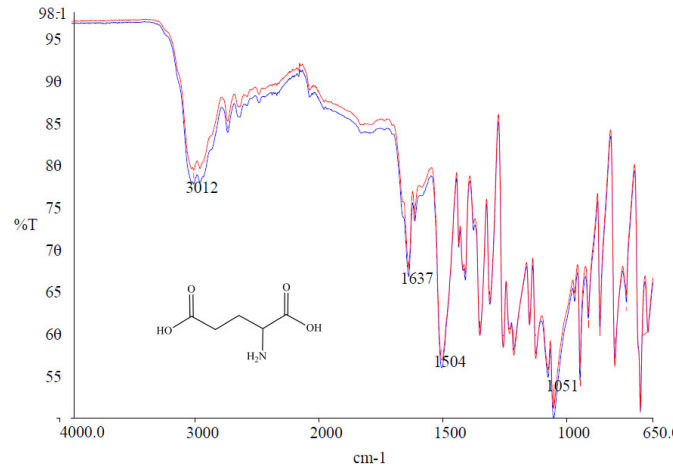

Figure 5: IR spectra before and after irradiated a sample at 230 kGy, and 295 K.

to the N-H bond (3). Finally, the other is at 1051 cm^{-1}, which is the vibration of the N-C (4). It is important to note that the same bands are observed after the analysis of the samples irradiated at 230 kGy at the three different irradiation temperatures, thus showing the stability of the LGA against ionizing radiation (Figure 6).

At the dose range and temperatures under study, glutamic acid is stable and forms very stable free radicals that can be quantified by Figure 6.

Two materials were analyzed that could serve as a support for the dosimeter: (A) PET, polyethylene terephthalate ($(C_{10}H_8O_4)_n$); and (B) the commercial paraffin ($C_{25}H_{52}$). The materials were irradiated at 230.4 kGy and at room temperature and were analyzed by ESR. The spectra do not show

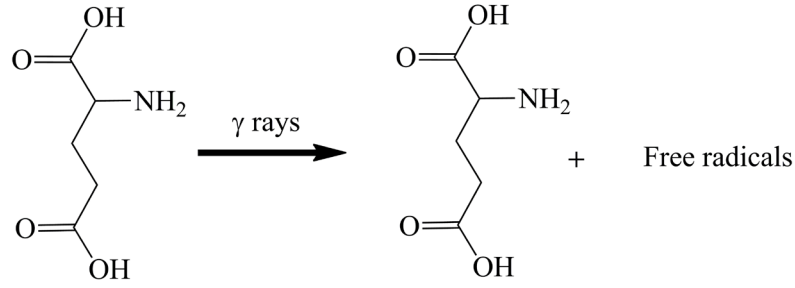

Figure 6: The results obtained suggested that glutamic acid (in a solid state) is a very stable molecule under ionizing radiation, and also the radicals that were produced.

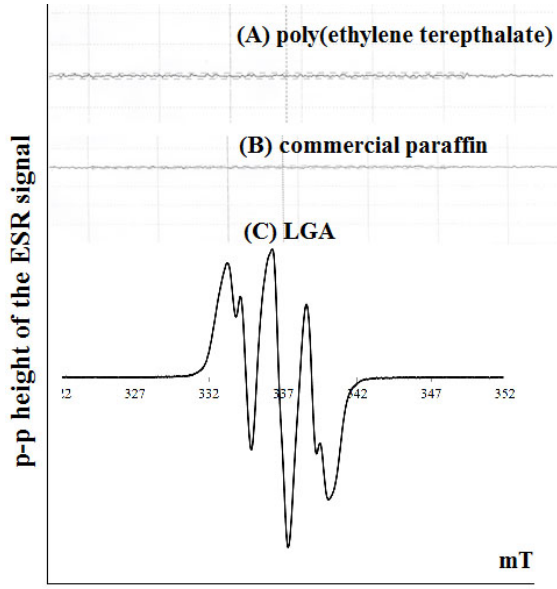

Figure 7. ESR analysis after irradiated samples of PET, paraffin, and LGA at 230 kGy and 295 K.

any significant peak that could interfere with the analysis in the interest range (Figure 7); therefore, these materials could be used as support of the glutamic acid. Besides, they are cheap and malleable.

REMARKS

We studied the response of the L-glutamic acid exposed to gamma irradiation at room temperature (295 K) and at low temperatures (77 and 195 K). When irradiation takes place at 77 K and 195 K, the concentration of the resulting radicals is lower compared with the concentration of the radicals formed at the higher temperature (295 K). The dose response curve showed linear behavior at all three temperatures from 43 to 230 kGy. These results are highly reproducible because the radicals formed are stable species. The radicals, formed after gamma radiolysis, are produced by decarboxylation and deamination reactions. ATR-FTIR analysis shows the stability of the amino acid under gamma rays, which is a result that may be of interest in other disciplines. With these preliminary experiments, we conclude that L-glutamic acid can be proposed for gamma dosimetry. Its advantages over other dosimeters are that L-glutamic acid is easily handled, and inexpensive. Experiments of absorbed

dose rate dependence, sensitivity, fading energy dependence, and lower limit of detection studies are under way.

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