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EFFECT OF GAMMA RADIATION ON SOME DOSIMETRIC INTEREST COMPOUNDS: AN EPR STUDY

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ABSTRACT. The present research was undertaken in order to investigate if some compounds used as natural sweeteners (steviol and sucrose) can be used as EPR dosimeters. The samples were exposed to low doses of gamma ionizing radiation used in the practices of radiodiagnostic medicine and interventional radiology. The EPR spectra of the gamma irradiated compounds reveal large signals, with multiple lines, which are characteristic for the free radicals in the solid state. From the analysis of the dependence of EPR signal of the absorbed dose, it was observed that by γ-irradiation the amount of generated radicals shows a linear dependence, which mean that there exist a possibility to use these two compounds as EPR dosimeters.

Keywords: *EPR dosimeters, γ radiation, free radicals, steviol, sucrose*

INTRODUCTION

Electron paramagnetic resonance (EPR) is a spectroscopic technique widely used in biology, chemistry, medicine, geology, materials sciences, antropology, physics in order to study systems with one or more unpaired electrons [1-5]. EPR spectroscopy has matureted into a powerful, versatile, nondestructive and nonintrusive analytical method. It was used as an investigative tool for the study of free radicals formed in the materials, since the radicals typically produce an unpaired spin on the molecule from which an electron is removed [6,7].

Also, the EPR spectroscopy has the ability to measure radiation-induced paramagnetic species, which persist in certain tissues (e.g., teeth,

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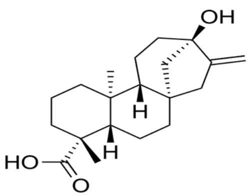
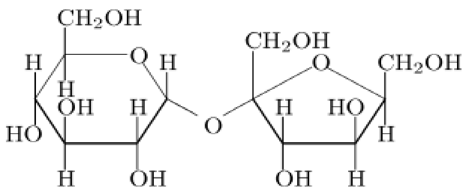
fingernails, toenails, bone, and hair). This fact has led that this technique to become an important method for screening significantly radiation-exposed individuals, by using dosimeters [8-10]. EPR dosimetry is based on the quantitative detection of stable paramagnetic species induced by radiation as a direct result of interactions with molecules, measuring the doses of energy absorbed by a matter, necessary to make the transitions, between energy levels of electrons.

In order for a material to be used as a dosimeter, it must meet certain well-defined parameters, for example: EPR signal with few lines, narrow and without signal in case of non-irradiation; stable radicals produced by ionizing radiation under normal conditions; low dose rate at low doses; high chemical radiative efficiency; adequate microwave power saturation properties allowing high power values for increased sensitivity; robustness of the dosimeter in terms of mechanical stress. The valorization and optimization of these parameters are the main goals of EPR dosimetry technologies, the studies being focused on the highest possible efficiency of the devices and techniques used [11].

Natural and artificial sweeteners as sucrose, steviol, xylitol, eritriol, aspartame etc., have been studied over time for their possible use as EPR dosimeters [12-14]. These compounds meet most of the well-defined parameters, which make them eligible to be used as EPR dosimeters. One of the advantages of solid EPR dosimetry is the easy determination of absorbed dose and non-destructive character of readings permitting to store the dosimeter as a document for further estimation [15].



(a)



(b)

Figure 1. Molecular structure of: (a) steviol and (b) sucrose.

Steviol is a chemical compound found in plants in the form of glycosidic steviol. It is responsible for the sweet taste of the leaves of *Stevia Rebaudiana*, a plant originating in Central and South America. The general chemical formula of this compound is $C_{20}H_{30}O_3$, its molecular structure being represented in (Figure 1a). Stevioside and Rebaudioside A, the primary components, are glycosidically attached to the hydroxyl functions of steviol in the structure of stevia. This compound is used as a sugar substitute, especially in diabetics and people on carbohydrate-controlled diets because it does not induce a glycemic response to ingestion.

Sucrose (Figure 1b), with the general molecular formula C12H22O11, is common sugar. It is a disaccharide, a molecule composed of two monosaccharides: glucose and fructose connected via a glycosidic bond. This type of linking of two monosaccharides is called glycosidic linkage. This is an important process for the storage and compression of energy. In this way, via sucrose, the plants can transport much easier large amounts of energy [16]. Sucrose has a monoclinic crystal structure and is quite soluble in water. Sucrose is produced naturally in plants, from which table sugar is refined.

The present research was undertaken in order to investigate if this two natural sweeteners, steviol and sucrose, can be used as EPR dosimeters. The samples were exposed to low doses of γ-ionizing radiation used in the practices of radiodiagnostic medicine and interventional radiology. The EPR spectra of the γ-irradiated compounds reveal large signals, with multiple lines, which are characteristic for the free radicals in the solid state [17]. From the analysis of the dependence of EPR signal of the absorbed dose, it was observed that by γ - irradiation the amount of generated radicals shows a linear dependence, which mean that there exist a possibility to use these two compunds as EPR dosimeters.

RESULTS AND DISCUSSION

Unirradiated and γ-irradiated samples of steviol and sucrose were studied by EPR spectroscopy. The EPR spectra of the irradiated samples are represented in (Figure 2). The unirradiated samples give no detectable EPR signal. The fact that non-irradiated samples show no EPR signal is a first condition accomplished by steviol and sucrose to be used as EPR dosimeters [11].

After irradiation both samples showed EPR signals, even at low absorbed doses, indicating a sensitivity to gamma irradiation of these compounds. The shape of the radiation-induced EPR spectra is similar to those described in the literature and has been attributed to the overlapping spectra of some radical species present in the samples. [14, 15, 19]. Large signals, characteristic for free radicals in the solid state, are present in the EPR spectra of low doses γ-irradiated samples. As it can be observed from (Figure 2), these EPR spectra consist of a few lines which are characteristic for the presence of free radicals in carbohydrate compounds [17, 18].

Also, the EPR spectra of the studied samples, recorded at X-band frequency (~ 9 GHz) at room temperature, due to broadened overlapping hyperfine lines of the different species, are unresolved and its cannot provide information about the radicals giving rise to its features [19].

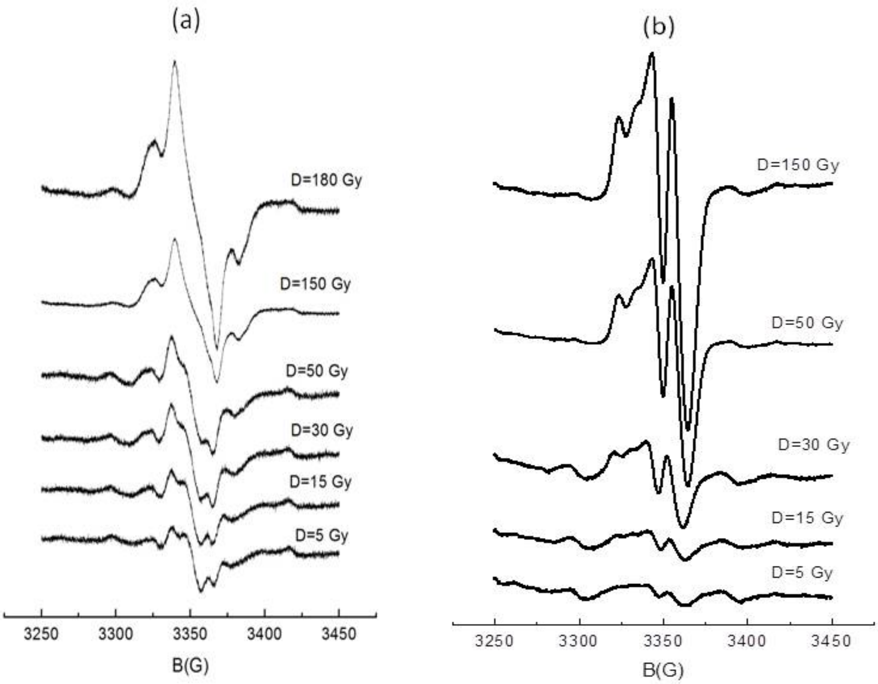
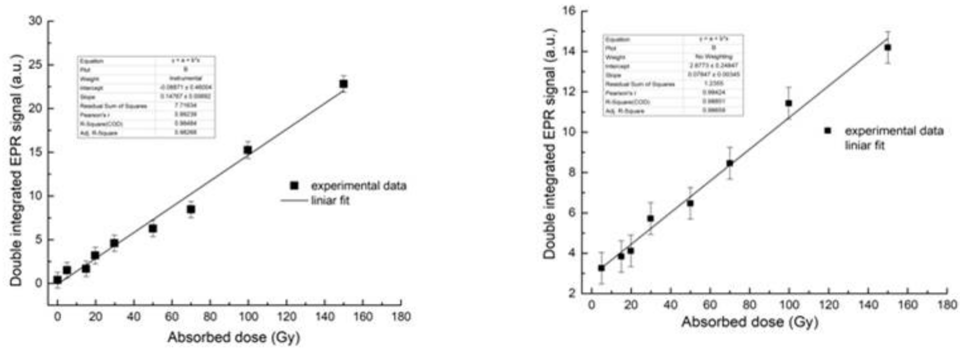


Figure 2. EPR spectra of γ-irradiated: (a) steviol and (b) sucrose, at different absorbed doses.

In (Figure 3) is represented the dependence of EPR signal intensities on the absorbed dose for steviol and sucrose. In order to generate the dose-response curve for steviol and sucrose, samples irradiated to 5, 15, 20, 30, 50, 70, 100 and 150 Gy absorbed doses were used.

A linear increase of EPR signal intensity is observed in both cases. The relative errors for all measurements were about 6%. The condition of linearity between dose and response is an essential one in biodosimetry, and the obtained results indicate a real possibility of using the studied chemical compounds in the determination of radiation doses.

Another important dosimetric property of a dosimeter is the stability over time of the radicals produced by ionizing radiation. It is an essential condition, because the measurement and estimation of the doses absorbed by the exposed persons is not done immediately [20]. There are studies in the literature that show that the intensity of EPR signals in stevia and sucrose samples decreases slightly over time [14, 21].



(a) (b)

Figure 3. Dose response relationship for (a) steviol and (b) sucrose as a function of the irradiated dose (errors bars are provided with 6% error limits).

In the present study, the effect of free radical recombination was studied by measuring the samples at room temperature at different moments of time after irradiation, concluding that this effect is insignificant [22,23]. This means that these substances are eligible for identification of the irradiation process even after a period of time from the irradiation. Therefore, the stability condition of the radicals produced by the ionizing radiation in the studied samples is achieved.

One of the important experimental parameters in evaluating the intensity of EPR signals is the microwave power. The intensity of an EPR signal increases with the square root of the microwave power in the absence of saturation effects. The effect of microwave saturation leads to a decrease in the amplitude of the EPR line with an increase in microwave power due to the reversal of the distribution of unpaired electrons on energy levels [24]. In order to find optimal nominal power settings, it is necessary to determine the microwave power saturation curves for the spectra to ensure that nonsaturating conditions were being used in the representation of the dose response curve (Figure 4). From (Figure 4) it can be seen that the intensity of EPR spectra of irradiated samples depend on the applied microwave power [14, 25].

Based on the microwave saturation, it was found that, in the case of the steviol sample, the intensity of the EPR signal increases suddenly until 5 mW, between 5 mW and 9 mW a slow increase is observed, and over 9 mW a steady plateau is reached, but the EPR signal it is not completely saturated (Figure 4a). Furthermore, it is obvious that the intensity of the sucrose EPR signal increases linearly with the increase of microwave power from 0.25 mW to 9 mW (Figure 4b). Even above 9 mW, the intensity of the EPR signal continues to increase, and above 16 mW the increase of the intensity is significantly lower.

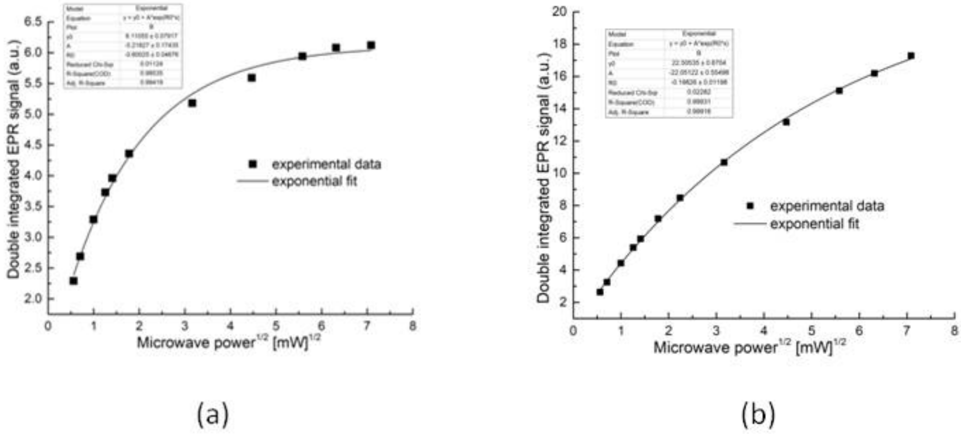


Figure 4. EPR power saturation curves for of irradiated steviol (a) and sucrose (b)

Continuous microwave saturation of EPR spectra indicated that faster spin-lattice relaxation-process existed in steviol sample than in sucrose sample (Figure 5).

It is obvious that the EPR line intensity reaches a maximum, much faster in the case of steviol than sucrose. That point in which the EPR line reaches a maximum is assigned as the "saturation point" based on the magnitude of the microwave power.

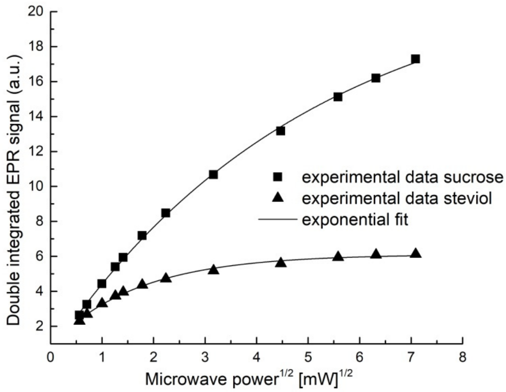


Figure 5. Comparation of EPR signal intensity irradiated samples.

CONCLUSIONS

EPR dosimetry is proving to be a method increasingly used in radiodiagnostics and interventional radiology, and as a result various compounds are studied to provide information "In vivo." The present research was undertaken in order to investigate if steviol and sucrose natural sweeteners, can be used as EPR dosimeter materials. The samples were exposed to low doses of γ-ionizing radiation used in the practices of radiodiagnostic medicine and interventional radiology.

A set of well-defined parameters specific to a material used as an EPR dosimeter was studied. From the results obtained, and following their analysis, it was found that the studied samples largely meet the requirements imposed on such a material. The non-irradiated samples show no EPR signal, and this is a first condition accomplished by steviol and sucrose to be used as EPR dosimeters. Also, the condition of linearity between dose and response is an essential one in biodosimetry, and the obtained results indicate a real possibility of using the studied chemical compounds in the determination of radiation doses. The stability condition of the radicals produced by the ionizing radiation in the studied samples it is achieved too.

Continuous microwave saturation of EPR spectra shows linearity between the intensity of the RES signal and the square root of the microwave power up to 5 mW for steviol, and up to 16 mW for sucrose, respectively (Figure 5).

EXPERIMENTAL SECTION

The samples with natural sweeteners, in polycrystalline form, were exposed to the action of low doses of gamma radiation generated by a ^{60}Co source (Gamma Chamber 600 from the Faculty of Physics) with a flow rate of D = 1.96 Gy/h, at different absorbed doses (from 5 Gy to 180 Gy). EPR measurements were performed using a Bruker EMX type EPR spectrometer operating in the X band (~ 9 GHz) with a modulation frequency of 100 kHz at room temperature. The amount of free radicals generated at different absorbed doses was assessed using the integrated and normalized double EPR signal per milligram of sample [17, 18], which is correlated with the number of paramagnetic species present in the sample. The integral intensities of the EPR spectra were obtained by evaluating their double integrals using the OriginPro2015 software.

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