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New ESR Dosimeter Materials Using Lithium Organic Acid Salts

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1. Introduction

Electron spin resonance (ESR) dosimetry using the signal intensity of radicals produced by radiation has been extensively studied in various fields. The dosimeters using organic substances are tissue-equivalent in the response to radiation energies and have generally a linear dose response over a wide enough range of dose to cover low and high doses. One of the advantages of ESR dosimetry is that the readout does not affect the spin concentration; the sample can be evaluated many times. Therefore, the signal to noise ratio (*S/N*) can be improved by repeated reading of the sample (Ikeya, 1993). Dosimetric applications of ESR have been successfully made for intermediate and high dose ranges using alanine as a dosimetric material (Ikeya, 1993; Kojima et al., 1986; Regulla & Deffner, 1981). Efforts to use ESR dosimeter in health physics have resulted in a minimum detectable dose of 10 mGy for sugar and 100 mGy for Li and Mg-Lactate (Nakajima, 1988; 1995; Nakajima & Otsuki, 1990; Hassan & Ikeya, 1997; Hassan et al. 1998; Hassan et al., 1999abc).

Although the sensitivity of a compact ESR dosimeter reader is being improved, finding a new material with higher sensitivity than alanine should lead to further advancement of ESR dosimetry. The new material should have a large *G*-value (stable radical pairs per 100 eV of radiation energy), a sharp linewidth and thermal stability at room temperature. It should consist of light elements in the periodic table so that the response does not depend on radiation energy (tissue equivalent). In contrast, the ionization efficiency of heavy elements depends on radiation energy since inner-shell ionization plays a significant role in radical formation by ionizing radiation (Ikeya, 1993).

Organic radicals, which are characteristic of the constituent molecules, are formed in a Van der Waals bonded molecular crystal (Ikeya, 1996). However, the linewidth is large due to superhyperfine (shf) interaction with nearby protons in neighboring molecules. If metal ions are situated between molecular ions, the linewidth might become small due to the reduction of shf interactions. A new material based on this conceptual idea is a metal ion-organic acid compound. The ionic bonding will increase the band gap, and intentional doping with aliovalent cations might enhance the sensitivity, as is commonly done for sensitization of the thermoluminescence dosimeter (TLD). A series of ESR dosimeters using organic ionic compounds, as a hybrid between metal ion and organic material, have recently been developed (Hassan & Ikeya, 1997; Hassan et al, 1998; Hassan et al., 1999abc). The requirement of a tissue equivalent dosimeter led us to use Li and Mg compounds rather than heavy alkali or alkali earth ions as the metallic cations. Li and Mg organic acid salts satisfied the dosimetrical and practical requirements, and are much more sensitive than alanine (Hassan et al., 1999abc; Hassan & Ikeya, 2000; Ikeya et al., 2000; Hassan et al., 2000). In this study, the possibility of using the new materials as ESR dosimeter materials is examined. The required properties of these materials for use as ESR dosimeter elements, such as dose response, annealing and sensitivity to ambient light, have been studied. Molecular orbital (MO) calculations of radical species have been performed to determine optical properties and ESR parameters, using commercial software.

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2. Results and Discussion

2.1. Spectral analysis of irradiated citrate

The ESR spectrum of Li-citrate is characterized by a main doublet signal which splits further into several more lines. The powder spectra of Li-citrate is so complex that we have calculated the MO of most conceivable radical species and then analyzed the spectra by choosing appropriate radical species.

2.1.1. Radical species from Li-citrate

Finch et al. (1979) found that three radical species were formed in a similar citric acid material after irradiation. Radical-A was the oxidized species produced by decarboxylation of the central carboxyl group with four β-hydrogen couplings. Radical-B was due to the abstraction of the hydrogen atom of CH_2, which displayed a doublet splitting due to one α-proton. Radical-C was the reduced species produced when an electron is added to one of the end carboxyl groups. Radicals-A and B were stable at room temperature while radical-C was not.

The radical species of Li-citrate were considered using models of radicals in citric acid. It was assumed that the homolytic cleavage of the central carboxyl group (radical-A), hydrogen abstraction of the CH_2 group (radical-B) and the reduced species are produced when an electron is added to one end of the carboxyl group (radical-C), as shown in Fig. 1(c).

Question 1 - What is one advantage of ESR dosimetry mentioned in the text?
A. It requires only one readout of the sample.
B. The readout does not change the spin concentration.
C. It can measure only low doses.
D. It works only with inorganic materials.

Question 2 - Which statement best explains why ESR dosimeters made from organic substances are suitable for a wide range of radiation studies?
A. They allow only a single measurement, reducing uncertainty.
B. They are non-reactive and therefore do not produce radicals when irradiated.
C. They respond to different radiation energies similarly to human tissue and show linearity over a broad dose range.
D. They can detect doses below 1 mGy without special preparation.

Question 3 - Based on the text, what does the improvement of the signal-to-noise ratio (S/N) in ESR dosimetry primarily depend on?
A. Increasing the radiation dose applied to the dosimeter
B. Repeatedly reading the same sample, since readout does not alter spin concentration
C. Using only inorganic dosimetric materials such as Li and Mg-Lactate
D. Cooling the sample to very low temperatures during measurement

Question 4 - Why must the new ESR dosimetric material be composed of light elements?
A. Light elements increase the rate of inner-shell ionization, improving sensitivity.
B. Light elements ensure that dose response becomes independent of radiation energy.
C. Heavy elements enhance thermal stability, which should be avoided.
D. Light elements naturally produce sharper ESR linewidths than heavy elements.

Question 5 - According to the text, which property is *not* explicitly required for a new high-sensitivity ESR dosimetric material?
A. A large G-value.
B. Thermal stability at room temperature.
C. Compatibility with heavy-element matrices.
D. A sharp ESR linewidth.

Question 6 - What is the main reason heavy elements are unsuitable for achieving tissue-equivalent ESR responses?
A. They have poor stability at room temperature.
B. They form unstable radicals that decay too quickly.
C. Their ionization efficiency varies with radiation energy due to inner-shell processes.
D. They produce excessively large G-values, causing nonlinear dose response.

Question 7 - Why might the incorporation of metal ions between molecular ions lead to a narrower ESR linewidth?
A. Metal ions increase the number of protons interacting with the radicals.
B. Metal ions reduce superhyperfine interactions by distancing nearby protons.
C. Metal ions decrease the band gap and thus suppress proton coupling.
D. Metal ions eliminate all hyperfine interactions altogether.

Question 8 - What is the primary reason Li and Mg compounds were chosen instead of heavier alkali or alkaline-earth ions in the development of new ESR dosimeters?
A. Li and Mg ions produce stronger ESR signals due to higher atomic numbers.
B. Heavy ions reduce sensitivity by producing broad ESR linewidths.
C. Li and Mg maintain tissue equivalence because their ionization efficiency does not strongly depend on radiation energy.
D. Heavy ions are chemically incompatible with organic acid salts.

Question 9 - Why was it necessary to calculate the molecular orbitals (MO) of multiple possible radical species when analyzing the ESR spectrum of Li-citrate?
A. Because Li-citrate forms only one dominant radical species that must be confirmed computationally.
B. Because the powder ESR spectrum is highly complex, requiring comparison with theoretical spectra of different radicals to identify which species contribute to the observed splitting.
C. Because MO calculations eliminate the need for experimental ESR measurements altogether.
D. Because the ESR spectrum of Li-citrate shows no splitting unless MO calculations are performed.

Question 10 - Based on Finch et al. (1979), which factor most clearly distinguishes radical-C from radicals-A and B?
A. Radical-C involves hydrogen abstraction of the CH_2 group.
B. Radical-C is formed by decarboxylation of the central carboxyl group.
C. Radical-C is unstable at room temperature, while radicals-A and B remain stable.
D. Radical-C exhibits four β-hydrogen couplings in its ESR spectrum.