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Lanthanide doped alkaline metal sulphates as candidates for EPR dosimetry

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The applicability of lanthanide doped alkaline metal sulphates as a new type of dosimeters for EPR dosimetery of ionizing radiation has been investigated in an attempt to obtain a dosimeter with better characteristics than the commonly used alanine dosimeter. Irradiation of samples doped with different lanthanides (Y, Ln, Gd) showed that the best sensitivity is obtained using dosimeters doped with $Y_2(SO_4)_3$. Different procedures for manufacturing dosimeters were studied and an optimum procedure was established. The time stability of the EPR signal of the irradiated $Y_2(SO_4)_3$ dosimeter was investigated using a ^{15}N-PDT standard and no fading of the EPR signal was observed over at least two weeks. The dose dependence of alanine and $Y_2(SO_4)_3$ doped $K_3Na(SO_4)_2$ dosimeters irradiated in the range 20 Gy – 200 kGy was analysed using a combination of 1-hit and 2-hit mechanisms of free radicals creation.

Keywords: lanthanide doped $K_3Na(SO_4)_2$, electron paramagnetic resonance, dosimetry.

INTRODUCTION

The extensive use of ionizing radiation in industry (food processing, sterilization, *etc*.) and medicine has prompted the development of versatile and reliable dosimetric techniques capable of covering a wide range of doses (*e.g.,* 1 Gy–100 kGy). One of the methods currently employed is solid state EPR (electron paramagnetic resonance) dosimetry where the absorbed dose is assessed by measuring the concentration of free radicals induced in the sample by ionizing radiation. The SS EPR dosimetry has several favorable features desirable for any dosimetric system: small size, long lasting record (restricted diffusion prevents recombination of radicals), and the ability for repetitive dose determination due to the non-destructive nature of the read-out procedure. Dosimeters containing a mixture of alanine powder and binding substance have been proposed as the optimal material for EPR dosime-

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try[1] because of the following characteristics: reasonably high sensitivity, linear dose response in the range of 10 to 10^4–10^5 Gy, low costs, ability to be easily manufactured in any shape (films or rods), or size, and long-term stability (transfer dosimetry).[1–4] ALA-dosimeters are now commercially available and are routinely used in a wide range of facilities from radiation processing units to, for example, high-energy particle accelerators.[5]

In spite of their good characteristics, ALA-dosimeters show rather low sensitivity at low, radiotherapeutic doses (*i.e.,* around 2 Gy) and deviation from linear response for doses above 20–50 kGy. Therefore, investigations have been directed toward the search of materials that exhibit a higher *G*-value (the number of paramagnetic centers produces by the radiation per absorbed 100 eV) and/or materials that have EPR signal saturation above 0.3–0.5 MGy. Two types of materials are currently under investigation: a) substances similar to alanine such as ammonium tartrate[6] or Mg-lactate[7] and b) inorganic sulphates doped with various cations where the radiation induced SO_3^- radical is used for dose assessment.[8–10] We have investigated lanthanide doped alkaline metal sulphates (Ln-dosimeters) since it has been claimed that such substances exhibit extremely high sensitivity,[8] but also that similar types of material can be used for the assessment of very high doses.[9]

Film dosimeters give a lower EPR intensity than rod shaped dosimeters, because of the smaller amount of EPR active material within the most sensitive region of the EPR cavity. Nevertheless, we choose to investigate film dosimeters since many applications require determination of the spatial dose distribution, which can only be achieved by arranging film dosimeters into sandwiches. The investigations reported here are parts of a larger effort to develop a range of dosimeters that could be suitable for a wide range of irradiation conditions (medical or industrial, *i.e.,* different dose and dose rate ranges) as well as for different types of ionizing radiation (γ-rays, e^-, p^+).[11,12] Some preliminary results on lanthanide-doped dosimeters have been reported previously.[13]

EXPERIMENTAL

Preparation of dosimeters. Although ALA-dosimeters are now commercially available, our intention was to compare the dosimetric properties of ALA and Ln-doped $K_3Na(SO_4)_2$ dosimeters which are manufactured under same conditions. Therefore, alanine dosimeters were prepared by one of the routinely used procedures: polycristalline DL-α-alanine (BHD Chemicals Ltd.p.a.) and low-density polyethylene (PE16, Lotrene Cd0302) were mixed (50/50 % w/w) and grinded. A homogeneous mixture was then pressd-molded at 470 K into 0.3 thick plates, from which 5×10 mm dosimeters were cut.

The applicability of lanthanide doped $K_3Na(SO_4)_2$ as a dosimetric system was evaluated using the following preparations. The first step was to assess: a) which lanthanide ion as dopant yields the highest sensitivity of the doped $K_3Na(SO_4)_2$ matrix to irradiation; and b) whether it is necessary to add lanthanides as sulphates instead of the commercially available oxides to avoid an additional step in the dosimeter preparation. Three lanthanide cations (Y, La and Gd) were selected based on their efficiency as dopants.[8] Initially, the procedure outlined in Ohta *et al.,*[8] was used, *i.e.,* $K_3Na(SO_4)_2$ matrix was prepared by homogenizing Na_2SO_4 and K_2SO_4 in acetone, followed by heating to 340 K to evaporate acetone and then firing at 1023 K in an oven (Carbolite Furnaces) under

argon atmosphere for 4 h was adopted, lanthanide oxides or sulphates (dopant/matrix ratios: 0.074 mol % for Y, or 0.15 mol % for La or Gd were sected to yield maximum sensitivity[8]) were homogenized with the $K_3Na(SO_4)_2$ matrix in acetone and the entire procedure was repeated (*preparation I*).

After selecting the best candidate (*i.e.,* $Y_2(SO_4)_3$ for Ln-doped $K_3Na(SO_4)_2$ dosimeters, the optimal procedure of doping the $K_3Na(SO_4)_2$ with $Y_2(SO_4)_3$ was then evalualed. In addition to the previously described procedure, two other preparation methods were employed: a) $Y_2(SO_4)_3$ and Na_2SO_4/K_2SO_4 were mixed in acetone and heated only to 340 K (*preparation II*), and b) such a mixture was fired at 1023 K in an oven under argon atmosphere for 4 h (*preparation III*), *i.e.,* the preparation of a fired matrix was avoided. The ground powders were then irradiated and their EPR spectra recorded shortly after irradiation (Fig. 2b, Tables I and II).

Finally, Y-film-dosimeters were prepared by grinding the mixture (60:40 % w/w) of $Y_2(SO_4)_3$–$K_3Na(SO_4)_2$ powder (prepared by *preparation III*) and polyethylene which was then press-molded to produce 0.3 mm thick films from which 5×10 mm dosimeters were cut (*i.e.,* identical shape as for the ALA-dosimeters). Both types of dosimeters were irradiated using a wide range of doses and their responses to irradiation were compared. The dosimeters were kept at ambient temperature and humidity.

Irradiation. ^{60}Co γ-ray irradiation was performed at the Institute of Nuclear Sciences "Vinča", Belgrade, using either a low dose rate source (≅ 100 Gy/h) for the dose range 10 – 200 Gy or a high dose rate source (1.20 kGy/h) for the dose range 0.2 – 200 kGy. The dosimeters were irradiated under electronic equilibrium at room temperature.

Preparation of standard. Nitroxide ^{15}N-PDT (2,2,6,6-tetramethylenpiperidine-d_{16}-^{15}N-oxyl-4--one, MSD isotopes, Los Angeles, CA) was used as an external standard in order to improve the reproducibility of the EPR measurements of the dosimeters. The reason for using perdeuterated, ^{15}N substituted nitroxide was its feature to exhibit two very narrow EPR lines[14] (instead of the 3-line spectrum of ordinary ^{14}N nitroxides) which do not overlap with the line of the dosimeter (see Fig. 3). The nitroxide was dissolved in distilled water (concentration ≅ 0.3 mM) and the solution was drawn into a capillary, both ends of which were consequently sealed. The standard was then fitted into the EPR holder (Fig. 1) and used to check the stability of the EPR spectrometer as well as in experiments where the fading characteristics of the Y-dosimeters were investigated (see Fig. 4).

EPR measurements. All EPR spectra were recorded at room temperature using a Varian E104-A or E-109 EPR spectrometer operating at X-band (9.5 GHz). The modulation frequency was 100 kHz and other scanning parameters (*i.e.,* modulation amplitude, *MA*; microwave power, *P*; and scan range) were selected on the basis of detailed assessment of the behavior of the examined EPR species (see Fig. 5). EPR spectroscopy is notoriously unreliable for quantitative measurements due to the inhomogeneous magnetic field within the cavity. Cosequently, the positioning of the samples is of vital importance[15] and all dosimeters were measured within the special holder shown in Fig. 1. The holder enables precise and reproducible positioning of dosimeter film vertically and parallel to the magnetic field. In addition, it enables the insertion of the external standard, which was used in some experiments (see Figs. 3,4). The dose response of dosimeters was calculated from the measured peak-to-peak height of the first-derivative of the central line of the EPR spectra, normalized to the unit weight of dosimeter and the receiver gain.

Data analysis. EPR spectra were recorded and analyzed using EW software (Scientific Software). The dose dependence of both the ALA- and Y-dosimeters (the EPR signal *vs*. absorbed dose) was analyzed using the formalism previously developed for similar types of dosimeters.[10,16] Data on the dose response (Fig. 6) were fitted using the Table Curve 2D program.

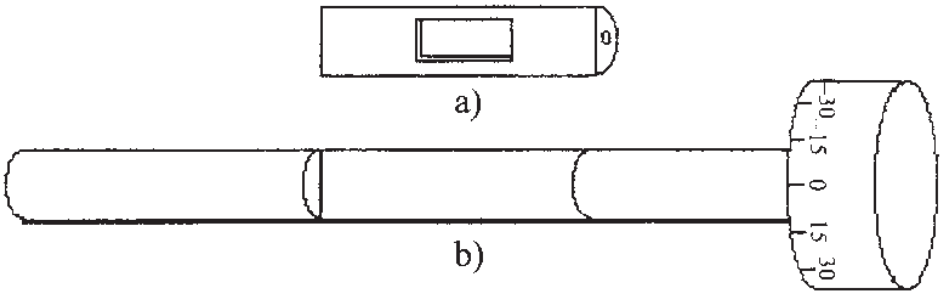

Fig. 1. A sketch of the specially designed EPR sample holder. Film dosimeters were placed into the rectangular hole, while the cylindrical hole is for the placement of standards.

RESULTS AND DISCUSSION

I. EPR spectral characteristics of irradiated Ln-doped $K_3Na(SO_4)_2$

The EPR spectrum of the ALA-dosimeter (Fig. 2a) shows the well known five line spectrum (intensity distribution is approximately 1:4:6:4:1) of the $CH_3C\dot{H}COOH$ free radical, resulting from the loss of the NH_2 group.[1,3] The EPR spectra of Ln-doped $K_3Na(SO_4)_2$ show only a single line with a width of around 1 mT. The type of preparation or the type of dopant (Y or La) did not have any influence on the shape of EPR spectra, but only on their intensity. It is also noteworthy that irradiation of undoped $K_3Na(SO_4)_2$ gave an EPR signal identical to Ln-doped $K_3Na(SO_4)_2$ (Fig. 2b). An EPR signal at zero dose was not detectable in any of the preparations. Measurement of the *g*-value for this radical, calculated from the known values for ^{15}N-PDT, gave $g = 2.0037$, which agrees well with the published *g*-values of the SO_3^- radical.[8–10] The EPR spectral characteristics of the irradiation induced species shown in Fig. 2b also agree well with the EPR spectra obtained for the SO_3^- radical using a similar dosimeter preparation.[8,9] Other radicals (*e.g.,* SO_4^-, O_3^-) can also be produced by irradiating solid alkaline (or earth alkaline) metal-SO_4 matrix, however, they have a much lower signal intensity and are EPR 'visible'only if the spectra are recorded at very low temperatures.[10]

II. The effect of preparation on the dose response of Ln-doped $K_3Na(SO_4)_2$

Tables I and II summarize some of the main features of the Ln-doped $K_3Na(SO_4)_2$ materials in regards to their response to irradiation. It is not surprising that irradiation of an undoped matrix gave a weak EPR signal as compared to the Ln doped one (Table I, Fig. 2b), since it has been found that undoped alkaline (or earth alkaline) metal-SO_4 matrix is not sensitive enough to be used for radiation dosimetry.[10] Also, it is obvious that using Y as the dopant provides a better dose sensitivity than using La. The EPR spectrum of Gd-doped $K_3Na(SO_4)_2$ was contaminated by a broad line (presumably from Gd) superimposed on the EPR line of the SO_3^- radical. It appears that adding the lanthanides as sulphates has advantages in regards to the dose response over lanthanide oxides (compare EPR signals for $Y_2(SO_4)_3$ and Y_2O_3) in spite of a rather low level of doping (0.074 mol %).

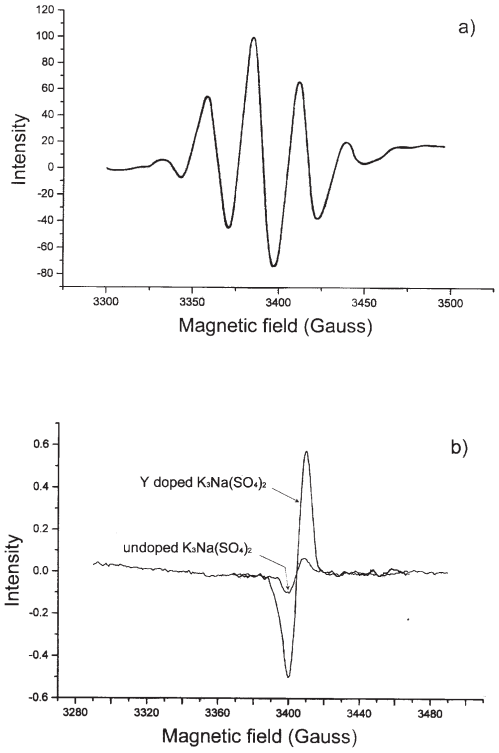

Fig. 2. Typical EPR spectra of a) alanine and b) Y-doped $K_3Na(SO_4)_2$ and undoped $K_3Na(SO_4)_2$ matrix irradiated with ^{60}Co γ-rays (100 Gy).

TABLE I. EPR signal intensity (arbitraty units, normalized to sample weight and instrumental gain) of irradiated $K_3Na(SO_4)_2$ matrix and Ln doped $K_3Na(SO_4)_2$ matrix. All samples were prepared using *procedure III* (see Experimental).

Sample	EPR signal intensity (a.u.)	
	20 Gy	100 Gy
Y(SO)KNa(SO)	79	267
YOKNa(SO)	44	136
LaOKNa(SO)	15	60
KNa(SO)-matrix	9	21

Table II shows that particular details in the preparation of Ln-doped $K_3Na(SO_4)_2$ dosimeters can have a profound influence on their response to irradiation. The high-temperature treatment in an inert atmosphere is obviously unavoidable (compare EPR signals for procedure II *vs*. procedures I and III). However, the best results were obtained by slurring $Y_2(SO_4)_3$ with Na_2SO_4/K_2SO_4, followed by firing, without previous preparation of the $K_3Na(SO_4)_2$ matrix as it has been recommended[8] (procedure III *vs*. procedure I). This appears to be consistent with the proposed mechanism according to which lanthanide dopants substitute Na in the $K_3Na(SO_4)_2$, thus promoting the formation of SO_3^- *via* charge compensation,[17] which in turn should favor procedure III.

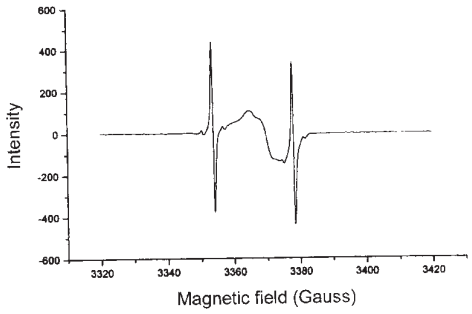

Fig. 3. The EPR spectra of SO_3^- free radical induced by irradiation of $Y_2(SO_4)_3$–$K_3Na(SO_4)_2$ film dosimeter and ^{15}N-PDT nitroxide standard measured in the EPR sample holder (see Fig. 1).

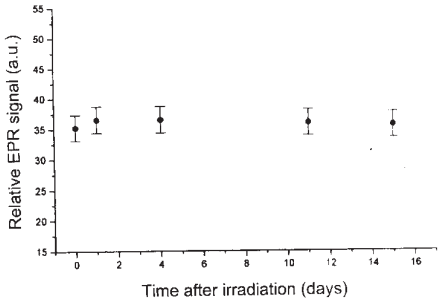

Fig. 4. Time stability of SO_3^- free radicals induced by irradiation of a $Y_2(SO_4)_3$-$K_3Na(SO_4)_2$ dosimeter. The EPR signal is given as the ratio of the EPR signal amplitudes of SO_3^- and the standard (see Fig. 3).

TABLE II. EPR signal intensity (arbitrary units, normalized to sample weight and instrumental gain) of irradiated $Y_2(SO_4)_3$ doped $K_3Na(SO_4)_2$ matrix prepared using different procedures (see Experimental)

Dose/Gy	Procedure I[8]	Procedure II	Procedure III
20	55	12	79
100	190	28	267

III. Stability of radical in Y-dosimeters

The fading of the EPR signal of irradiated dosimeters is an essential parameter in assessing of their practical applicability. If the signal fading is reproducible and well known, it can be corrected for, however, it is advantageous to have dosimeters the EPR signal of which is stable over a longer period of time (at least a few weeks after irradiation). We used the external standard to minimize errors arising from the spectrometer tuning and dosimeter positioning during repetitive measurements. Figure 3 shows the EPR spectrum of the Y-dosimeter placed into the sample holder along with the ^{15}N-PDT standard. The separation of the EPR lines of standard and dosimeter is sufficient for independent measurements of their respective amplitudes thus enabling the use of relative intensities and avoiding the nuances of EPR metrology. The Y-dosimeter was irradiated by 8 kGy and repeatedly measured over a period of 15 days (Fig. 4). The reproducibility of EPR measurements was assessed using the same irradiated Y-dosimeter and ^{15}N-PDT standard but taking out and reinserting the

holder into the cavity, while trying to reproduce the spectrometer tuning in each consecutive scan (error bars in Fig. 4). It is evident that irradiated Y-doped $K_3Na(SO_4)_2$ shows no detectable fading over the examined period (similar to ALA-dosimeters[18]) which makes it a good candidate for solid-state EPR dosimetry.

IV. Comparison of Y- and ALA-dosimeters

When comparing the characteristics of different dosimeters it is essential to determine the relationship between the EPR signal intensity (*SI*) and the instrumental parameters such as microwave power (*P*) and modulation amplitude (*MA*) for each dosimeter. An increase in both parameters increases the *S/N*-ratio, but on increasing *P* above a certain value, the *SI vs.* $P^{1/2}$ departs from linearity and eventually reaches a maximum (T_1 relaxation time effect), while increasing *MA* above a certain value results in line broadening and departure from linearity of *SI vs. MA*. The recommended *MA* and *P* values for ALA-dosimeters varies over a rather broad range (*MA* = 1 – 10 Gauss, *P* = 1 – 50 mW) which is the consequence of whether the authors kept these settings in the regions of linearity or attempted to increase the *S/N*-ratio while sacrificing the resolution of the spectrum structure. A systematic study of the EPR settings for dosimeters based on the measurement of SO_3^- radicals has not been reported. Therefore, to make a comparison between the Y– and ALA–dosimeters irradiated under same conditions it was necessary to determine the optimal values of *P* and *MA* for each dosimeter. Figure 5 shows the *SI* obtained at different settings of *MA* and *P* for the two dosimeters. The *SI* of the Y-dosimeters departs from linearity at a slightly higher *MA* than the ALA-dosimeter and also saturates at a slightly higher *P*.

Measurements of the signal intensities (*SI*) of the central EPR lines of the two types of dosimeters, which were irradiated over a broad range of absorbed doses and measured under identical instrumental conditions, indicate that the ALA-dosimeters are more sensitive than the Y-dosimeters (Fig. 6). Independent measurements of dosimeters irradiated with 10 kGy and recorded under *MA* = 1 Gauss and P = 5 mW give the ratio $SI_{ALA}/SI_Y = 1.62$. This ratio can be improved by increasing the values of *MA* and *P* when measuring Y-dosimeters, so eventually an equal sensitivity can be achieved. The value of the double integral (DI) of the EPR spectra is physically more meaningful when comparing the sensitivity of two dosimeters since this, in theory, should give the ratio of the absolute number of radiation induced radicals in each dosimeter. Unfortunately, this method has disadvantages since baseline fluctuation induces considerable uncertainties, so the peak-to-peak method appears to be the more reliable one. Nevertheless, the ratio of the double integrals under above conditions was 2.73, *i.e.,* higher than the ratio of *SI*'s, which is to be expected from the difference in the respective spectral shapes (Fig. 2). Although this is not relevant from a dosimetric point of view where only the peak-to-peak method is commonly used, this analysis is important since our results are inconsistent with the result published by a Japanese group[8,17] who reported extremely high sensitivity for the same type of dosimeters, *i.e.,* a change of the EPR signal intensity by almost one order of magnitude per 10 Gy and the ability to measure absorbed doses down to 0.1 mGy. This would imply that their dosimeters were about three orders of magnitude more sensitive than alanine dosimeters. Since the

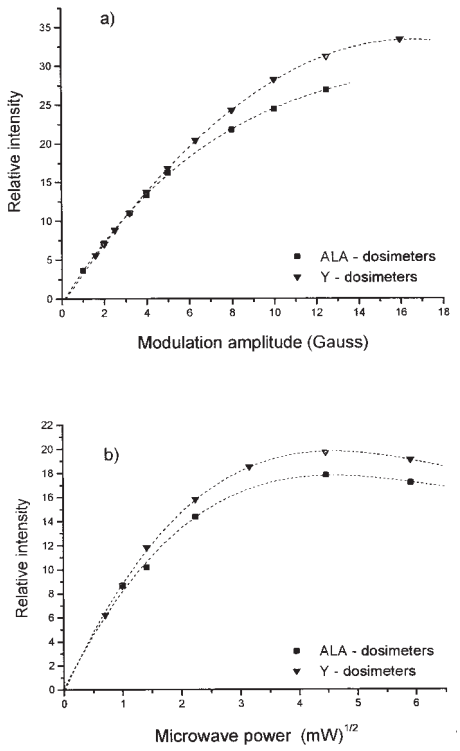

Fig. 5. Relative EPR signal intensities of irradiated ALA- and Y-dosimeters recorded under different instrumental settings a) *SI vs.* modulation amplitude (microwave power = 2 mW), b) *SI vs.* the square root of the microwave power (modulation amplitude = 3.2 G).

sensitivities of our ALA- and Y-dosimeters are similar to previously reported sensitivities for these types of dosimeters,[1,6,9] the possibility that a discrepancy of three orders of magnitude in sensitivity between our measurements and those by the Japanese group is due to the different sensitivity of the EPR spectrometers can be ruled out. In any case, the reported ability to perform measurements of absorbed doses in the 0.1 mGy range would require G-value of 100 or more, which seems rather unrealistic.

It is obvious that the shape of the dose response curves of the two dosimeters differ, *i.e.,* the Y-dosimeters show supralinearity, while the ALA-dosimeters show sublinearity (Fig. 6). We tried to decompose the dose-response curves in Fig. 6 using a theory proposed for both TLD and EPR dosimeters.[10,16,19] The measured experimental points were fitted assuming a mixture of 1- and 2-hit response using the following equation:[16]

$$SI(D) = R(1-\exp(-D/D_{01})) + (1-R)[1-(1+D/D_{02})\exp(-D/D_{02})]$$

where R is the relative contribution of 1-hit, and D_{01} and D_{02} are characteristic doses for 1- and 2-hit events. From the above equation it can be seen that 1-hit factor R characterizes the dose-response is sublinear for $1/2<R\leq 1$ and for $0\leq R<1/2$ suplralinear the dose-response is sublinear for $1/2<R\leq 1$ and for $0\leq R<1/2$ supralinear, while the D_0's can be used to compare the sensitivity and/or linearity of various

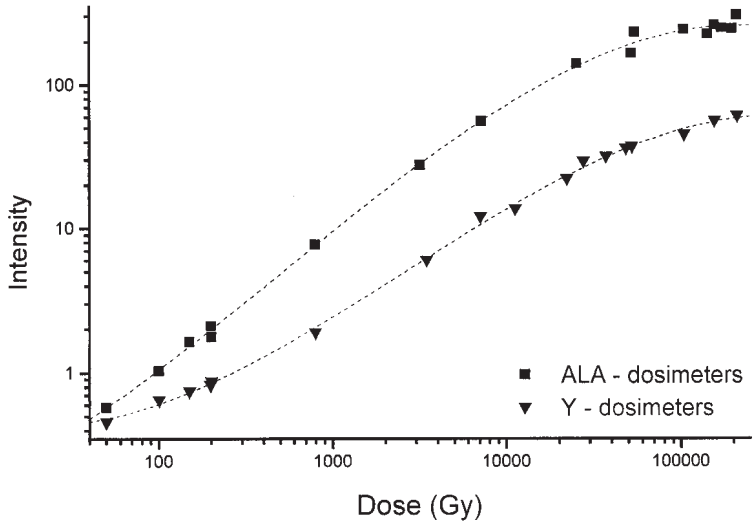

Fig. 6. EPR signal intensities of irradiated ALA- and Y- dosimeters (recorded under the same instrumental settings) *vs.* absorbed dose.

dose responses.[10] The decomposition into 1- and 2-hit parts is shown in Fig. 7. The best-fit parameters were: $R = 0.67$; $D_{01} = 22$ kGy; $D_{02} = 98$ kGy for ALA-dosimeters and $R = 0.33$; $D_{01} = 21$ kGy; $D_{02} = 103$ kGy for Y-dosimeters. The obtained values of R indicate the different contribution of 1- and 2-hit events between the two types of dosimeters, *i.e.,* the difference in the mechanism of interaction between the γ-rays and target media. The values of the D_0's are almost the same indicating that both dosimeters are able to cover the same range of doses prior to saturation. However, it is obvious from Fig. 7b that the (1+2)-hit model does not adequately describe the dose-response curve of Y-dosimeters and that inclusion of some additional mechanisms seems necessary.

CONCLUSIONS

This investigation demonstrated that Ln-doped $K_3Na(SO_4)_2$ are potentially useful materials for EPR dosimetry. From a spectroscopic point of view, their EPR spectra have favorable characteristics (*i.e.,* a single line spectrum). They also do not show EPR signal fading for at least 15 days, a time sufficient for accurate dose measurements, as well as for transfer dosimetry. These materials showed higher dose sensitivity than pure alkaline (or earth alkaline) metal-SO_4 which points to the importance of the nature of the doping agent, as well as to the particulars of the doping procedure. Although some authors have proposed an explanation for the doping mechanism,[17] our present knowledge is still at the empirical level, hence the search for the 'ideal' dosimeter might require information from additional techniques such as X-ray crystallography. Although lanthanide doped alkaline metal sulphates did not show better dose sensitivity than routine alanine dosimeters, EPR investigations of these materials are of interest because similar materials have been used for

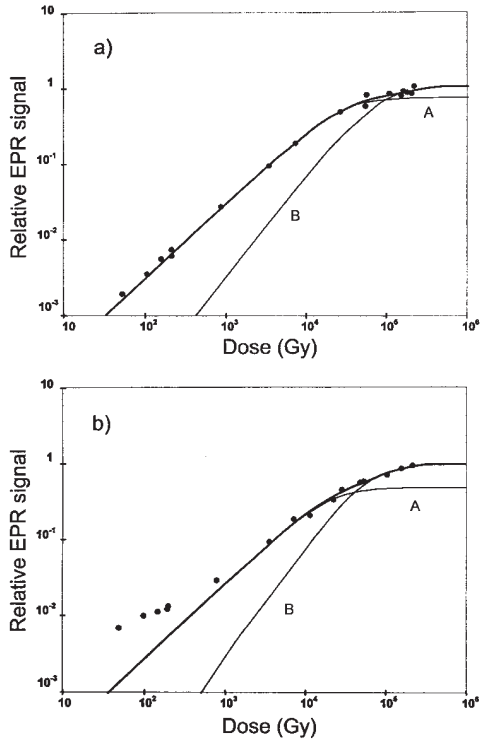

Fig. 7. The dose-response for ALA- and Y-dosimeters. Peak heights were normalized to saturation values. The thick lines throught experimental points are respective (1- and 2-hit) mixtures, while the thin lines show the individual components.

thermoluminescent (TLD) dosimetry. Therefore, EPR studies of these materials can contribute to a better understanding of the processes that occur in TLD dosimeters.

ИЗВОД

ЛАНТАНОИДИМА ДОПИРАНИ СУЛФАТИ АЛКАЛНИХ МЕТАЛА КАО КАНДИДАТИ ЗА ЕПР ДОЗИМЕТРИЈУ

ЈЕЛЕНА ПЕТКОВИЋ, ИВАНА МЛАДЕНОВИЋ, НИКОЛА ВУКЕЛИЋ, МИЛОШ МОЈОВИЋ и ГОРАН БАЧИЋ*

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Испитана је примена сулфата алкалних метала допираних лантаноидима као новог типа дозиметара за ЕПР дозиметрију јонизујућег зрачења у покушају добијања дозиметра који би имао боље особине од рутински употребљаваног аланинског дозиметра. Озрачивање узорака допираних са различитим лантаноидима (Y, Ln, Gd) показује да се најбоља осетљивост добија користећи дозиметре допиране $Y_2(SO_4)_3$. Испитани су различити поступци припреме дозиметара и утврђен је оптимални поступак. Коришћењем ^{15}N-PDT стандарда испитана је временска стабилност сигнала озраченог $Y_2(SO_4)_3$ дозиметра. Није примећено опадање ЕПР сигнала у току две недеље, што је довољно за трансфер дозиметрију. Анализирана је дозна зависност интензитета ЕПР сигнала аланинског и дозиметра на бази $K_3Na(SO_4)_2$ допираног $Y_2(SO_4)_3$ озрачених

у опсегу доза 20 Gy – 200 kGy коришћењем механизама комбинације једног и два поготка при стварању слободних радикала.

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