ALKALINE-EARTH METAL DITHIONATES-BASED SPIN-RESONANCE β -RADIATION DOSIMETERS

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Our previous studies have revealed that stable paramagnetic centres (PMC) classified as ion-radicals SO_3^- and characterized by the EPR isotropic signal (ΔH~5 G, g=2,0036) are formed on γ-irradiating the alkaline-earth metal dithionates of a general formula $MeS_2O_6 \cdot xH_2O$, where M is the alkaline-earth metal (1). Further studies gave us the reason to suggest barium dithionate ($BaS_2O_6 \cdot 2H_2O$) as a γ-dosimeter in the range of 10^{-1} to 10^4 Gy (2).

The study of ionizing radiation effect on the alkaline-earth metal dithionates has revealed also that this class of substances is not only γ-sensitive but β-sensitive as well. The literature on the subject cite only a few chemical compounds and materials used for the purposes of electron radiation dosimetry (3-6), e.g. L-alanine with the operating measurement range of 10^2 to $2 \cdot 10^5$ Gy (6). This paper describes a problem of dithionates application for the purposes of spin-resonance electron radiation dosimetry.

Barium and strontium dithionates were chosen as samples. The purity was tested by means of X-ray phase analysis, EPR technique and IR-spectroscopy. The preground salt powders were subjected to external and incorporated β-irradiation. External β-irradiation was carried out in the air at 20°C by the electron flow (10^{11} to 10^{14} cm^{-2}, E=4 MeV) and $^{90}Sr+^{90}Y$ sources of 10^4 and 10^5 Bq activity. Internal β-irradiation was performed by ^{90}Sr radionuclides of 10^3 and 10^4 Bq activity introduced into the substances structure during their recrystallization with $^{90}Sr(NO_3)_2$.

Ion-radicals SO_3^- were exclusively stabilized in the result of external β-irradiation of alkaline-earth metal dithionates by monoenergetic flows (E=4 MeV) and $^{90}Sr+^{90}Y$ radionuclides as well as in the case of γ-irradiation. The primary stage of homolytic decay of the excited $S_2O_6^{2-}$-anion S-S bond was believed to be the reason of radiation-induced SO_3^- formation. And this very, the so-

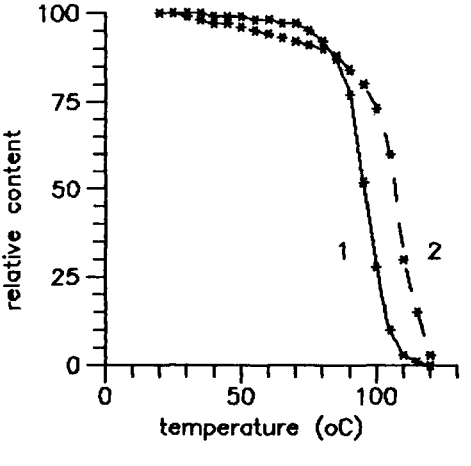

Figure 1. Crystallization water (1) and ion-radicals SO_3^- (2) content (%) vs temperature for $BaS_2O_6 \cdot 2H_2O$

called predissociative decay which essence is the preceding excitation energy migration and concentration on a particular bond, was suggested (7) for some (O-O, C-C, C-H) bonds, S-S bonds including.

Our studies have revealed that the process of stabilization of radiation-induced SO_3^- in the structure of dithionates was affected by the crystallization water. These PMCs were found to be very stable at 20°C. Nevertheless, even insignificant sample heating caused gradual recombination. Figure 1 (curve 2) shows the plot of SO_3^- relative concentration in $BaS_2O_6 \cdot 2H_2O$ structure versus temperature. This Figure shows insignificant annealing of radicals at about 90°C and their intensive recombination in the crystallization water removal region (curve 1). Noteworthy is that both processes terminate practically at a time. Besides, impossibility of dithionate original structure recovery by annealing SO_3^- was noticable. These observations had led to the conclusion that the processes of radicals recombination and crystallization water removal are interrelated. Such interrelation was also found out for radical particles stabilized in another non-organic systems, e.g. mixed crystals of $CaCO_3$ and $CaHPO_4 \cdot 2H_2O$ (8).

The comparison of external γ- and β-radiation effects reveals that the values of EPR signal intensity when exposing barium dithionate to equal doses of both kinds of radiation SO_3^- PMCs agree within the measurement accuracy (20%, 1σ) (Fig. 2). The intensity of EPR signal of

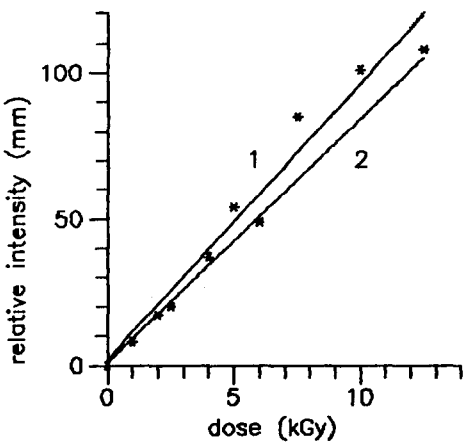

Figure 2. EPR signal relative intensity vs barium dithionate external gamma- (1) and beta-radiation (2)

SO_3^- was always a linear function both of β-radiation dose and exposure time for various activities of $^{90}Sr+^{90}Y$ sources used. Naturally, the angle of a straight line with the abscissa was changing. For example, the change of the source activity by one order (from 10^4 to 10^5 Bq) caused the increase of radiation-induced SO_3^- accumulation rate by about 7 or 8 time (Fig. 3).

The experimental data available made it possible now to preliminary estimate the external β-radiation absorbed dose by comparing the EPR signal intensities for γ- and β-radiation cases. Hence, the alkaline-earth metal dithionates may become promising spin-resonance dosimeters of β-radiation.

At present, large difficulties are experienced in β-radiation dose measurements for the case of incorporated radionuclides. We have attempted to compare the doses due to external electron and

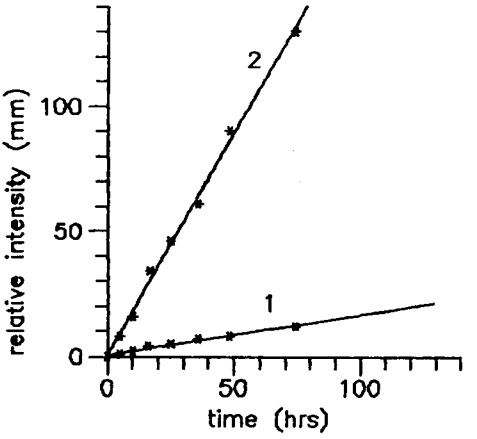

Figure 3. EPR signal relative intensity vs time. External β-irradiation of $BaS_2O_6 \cdot 2H_2O$ by $^{90}Sr+^{90}Y$ radionuclides of 10^4 Bq (1) and 10^5 Bq (2)

incorporated radiations. The intensity of a corresponding EPR signal was a working hypothesis when comparing the doses. When investigating crystal $BaS_2O_6 \cdot 2H_2O$ doped by $^{90}Sr+^{90}Y$ in the air at 20°C, the EPR signal was detected; this was a signal of radicals SO_3^- and its intensity was increasing linearly in time. Consequently, as it was the case of external β-radiation, it became possible to estimate the value of the absorbed dose of internal radiation by the very same method of compariny the number of radiation-induced PMCs respective to the applied dose of external γ- and internal β-radiation.

Thus the submitted material points out the below described advantages of alkaline-earth metal dithionates used as spin-resonance β-radiation dosimeters. Such advantages are high sensitivity to external and incorporated β-radiation, the narrow isotropic EPR signal for radiation-induced SO_3^-, large interval of linear accumulation, the PMC stability and discovered unsusceptibility to solar light. The instability at high temperature may be considered as a disadvantage of dosimeters described. Nevertheless, as it has been already pointed out, the ion-radicals SO_3^- are very stable at room temperature. The study of EPR signal intensity upon time has revealed that the intensity value is practically stable at temperatures below 40°C for at least 2 years (with ±10% accuracy (2σ)). This feature may provide for the lasting storage of information by sealed dithionate dosimeters.

REFERENCES

1. S.E.Bogushevich, V.N.Makatun, A.K.Potapovich and I.I.Ugolev, Zhurn. pricl. spectr. 55, 613-618 (1991).
2. S.E.Bogushevich, M.P.Lapkovsky, A.K.Potapovich and I.I.Ugolev, Avtor. cvidet. 1699267 SSSR, MKI ^{5} G 01 1/04 (1991).
3. Chen Wenxin, Jia Haishun, and Lii Gongxu, Radiat.Phys. and Chem. 33, 417-420 (1989).
4. K.Olsen, J.Hansen and M.Waligorski, Appl. Radiat. Isotop. A. 40, 985-988 (1989).
5. K.Mehta, R.Chu, and G. Van Dyk, Radiat. Phys. and Chem. 31, 425-434 (1988).
6. K. Van Laere, Appl. Radiat. and Isotop. A. 40, 885-895 (1989).
7. V.G.Plotnikov. 5th Work. Meet. Radiat. Interact, Leipzig 24 (1990).
8. T.Miki, A.Kai, Jap.J.Appl.Phys. 30, 404-410 (1991).