SOME PECULIARITIES AND COMPLICATIONS IN HIGH-DOSE ESR-DOSIMETRY

XA9949728

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Abstract

A number of experimental data reveal that ESR-dosimetry used for high doses sometimes fails as a result of radiation defect interactions at high radiation temperatures or after preliminary annealing stages, etc. Such interactions lead to non-linearities in the defect accumulation in a sample matrix vs. irradiation dose. The ways of accurate consideration of such effects by means of two phenomenological models are discussed.

1. INTRODUCTION

The ESR-dosimetry methods are very popular up to date, as they allow to determine, with a high accuracy and reproducibility, the integral radiation dose received by a particular person or some objects. Obvious advantages of the ESR-dosimetry are that it provides an opportunity to repeat measurements and to control the results obtained at various laboratories, because, when recording an ESR signal, the information contained in a sample is not lost and is conserved, practically, for a long time.

The advantages of the method are the results of it's physical essence: the magnetic resonance spectrum recorded without any lattice distortions (a working quantum $h\nu=\mu H<<kT$) serves for recording the number of radiation defects, used, in turn, for determining the radiation defect yield constant and, further, for calculating the dose absorbed by the sample.

Then it follows immediately that for correct application of the ESR-dosimetry methods, at least, two conditions are required. First, radiation defects in the sample used as a detector must be rather stable in order to have chance to neglect or to take into account accurately the influence of the recombination processes and other pathways of disappearance/transformation of paramagnetic centres (PMC). Secondly, one must know exactly the relationship governing radiation defects accumulation in a specific sample under some specified irradiation conditions. It is strongly desirable that this relationship is monotonous, i.e. it gives unambiguous predictions. Besides, stability of the defects relaxation characteristics is required, as variation in the magnetic relaxation time can result noticeably in the change of the spectrometer sensitivity.

2. RESULTS AND DISCUSSION

In practice, these conditions are satisfied quite well. The stability of the radiation defects and its relaxation characteristics are provided by an appropriate choice of the working sample material (for example, tooth enamel, crystal alanine, etc.). The relationship for radiation defect accumulation is determined by means of extra irradiations, and it can be considered as linear with the accuracy being sufficient in practice.

The situation, however, becomes more complicated if the conditions for defects interactions exit. Such conditions are present: at high radiation doses, at raised temperatures or mechanical pressure in a matrix during irradiation or at large dose rates when radiation heating of a sample is possible. In these cases, accurate determination of dose becomes more difficult; moreover, as it will be shown below, some ambiguities can arise.

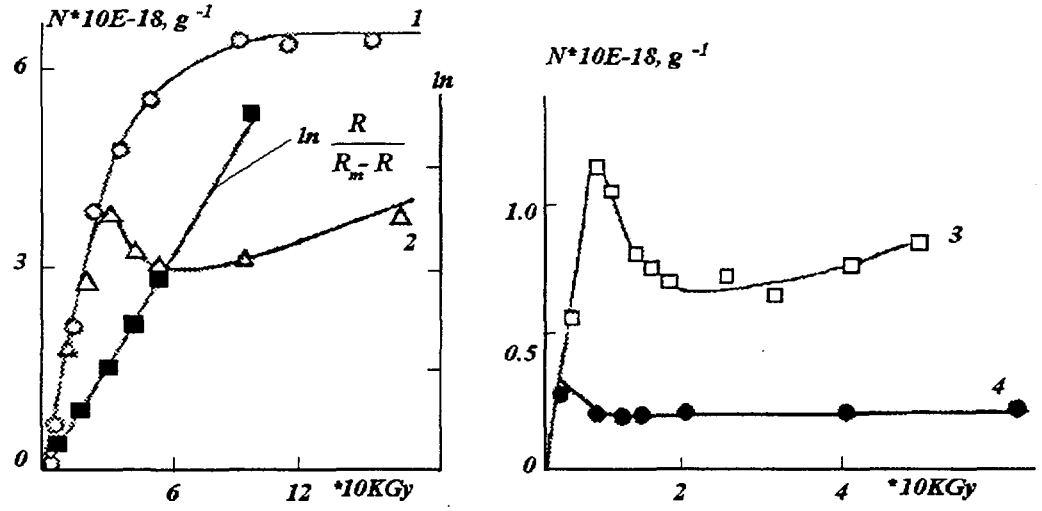


FIG. 1. Accumulation curves for free radicals in PMMA. 1) - at normal conditions (273 K); 2) , 3), 4) - at 305, 335, 381 K, respectively

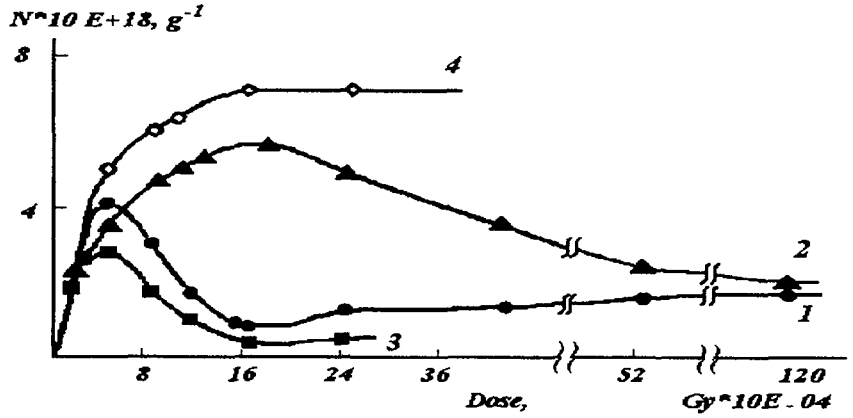


FIG.2. Accumulation curves for free radicals in PMMA. 1) - normal conditions; 2) - compressed (10 GPa); 3) - tensioned (25 MPa); 4) - low temperature, atmospheric pressure.

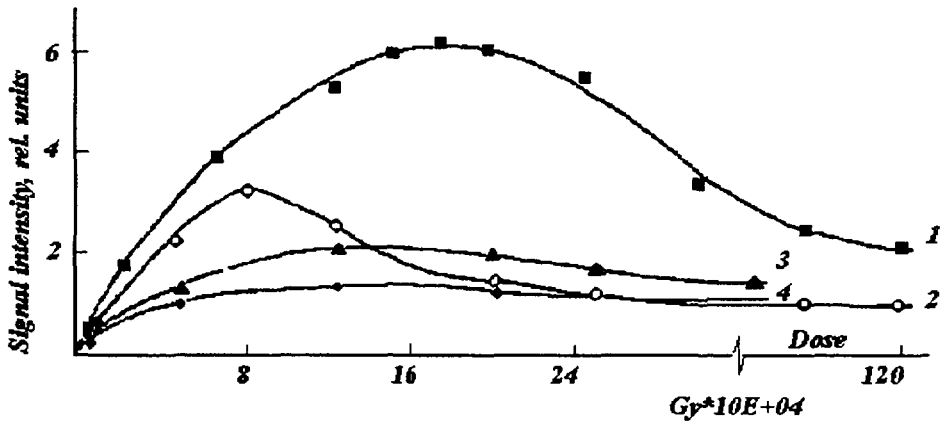


FIG. 3. Accumulation curves for free radicals in PMMA. 1) - initial irradiation at compression (10 GPa); 2) - initial irradiation at normal conditions; 3) - secondary irradiated under pressure after annealing; 4) - annealed and secondary irradiated without compression.

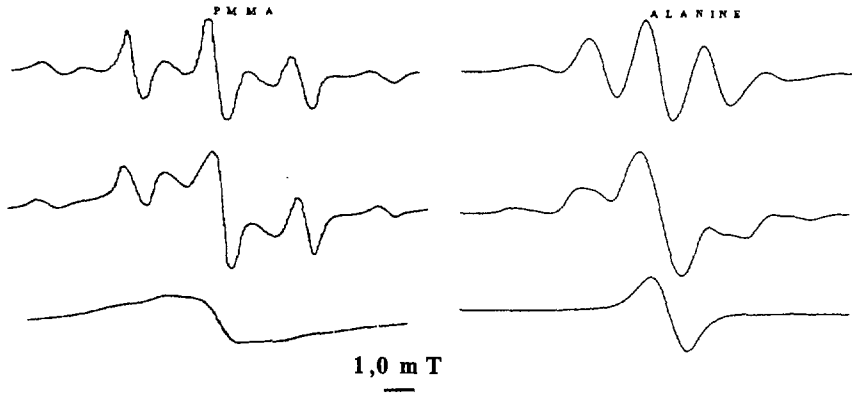


Fig. 4. The shapes of ESR signals in irradiated PMMA (left side) and α-alanine (right side) for low doses (top), intermediate doses or raised irradiation temperatures (middle), and very high doses or after annealing (bottom)

Another reason for troubles is a "radiation history" of a sample. Experiments have shown, that, as a rule, in dielectrics and in semiconductors the radiation yield constant strongly depends on whether the sample is irradiated for the first time, or it was earlier irradiated, annealed and then again irradiated. In the latter case in the course of repeated irradiations in many substances other types of defects, in addition to primary ones, are generated, therefore the relationship of PMC accumulation as well as its relaxation characteristics may change.

These concepts are exemplified below.

In Fig. 1, the curves of accumulation of radiation defects (in this case, radicals) in polymethylmethacrilate (PMMA) with irradiation dose are displayed for various irradiation temperatures [1].

As the sample temperature increases at various stages of irradiation, the accumulation curve shape varies significantly: it is transformed from an usual curve with saturation to a non-monotonic curve with extrema, thus for large doses an ambiguity occurs. Similar situation is observed [2] during irradiation of a sample at mechanical stresses (see Fig. 2). The compressing stress increases the maximum concentration of radicals and shifts the extremum to higher doses, while the tension causes an opposite effect. Like in the first case, these effects manifest only at rather high doses, whereas the specified peculiarities are, practically, non-distinguishable at small doses .

The "radiation history" of a sample plays an important role also. In Fig. 3 the accumulation curves of radiation defects in PMMA are shown for varied radiation conditions and "radiation history". One can observe a significant effect of preceding irradiation, annealing and mechanical pressure. The "radiation history" of a sample strongly influences also the amount of radiation defects that are stabilized in it.

PMMA is a more "soft" system, than alanine, which is the reasons why it is chosen as a main working sample for ESR-dosimetry; however, the same effects are observed in alanine too (see Fig. 4). Analogy with PMMA is evident, i.e. the transformation of the ESR spectrum that implies occurrence of other radicals, and the change in the PMC accumulation relationship as the radiation dose increases are observed.

In both cases, transformation of the PMC structure takes place: at the start of the radiation an alkyl type of radical stabilizes, and then an allyle one occurs. On the last steps, polyene radicals may appear [3].

For interpretation of these data and quantitative description of observable effects we have developed two phenomenological models, which take into account other types of defects that occur as the radiation dose increases (in this case allyle radicals are generated instead of alkyl ones). This can be a result of the interactions between the accumulated defects and the radiation field occuring as the dose increases.

This problem of the influence of the radiation field on the defects accumulation has been studied quite well. In numerous works (see references in [4]) it was shown that the interaction with a radiation field results in (beginning with a certain value of the dose) decreasing the number of stabilized defects and in saturating the accumulation curve. As a result, the *a priori* relationship of radiation defect accumulation

$$\frac{dN}{dD} = KD \tag{1}$$

changes to:

$$\frac{dN}{dD} = KD - K_1 N \tag{2}$$

where, N is the number of stabilized defects in a matrix, D is the irradiation dose, K is determined via the dose rate and the radiation yield of the defects, the constant K_1 takes into account the disappearance of the defects as a result of the radiation field influence.

In Ref. [4], it is stressed that Eq. (2) describes rather a large amount of observed experimental data (provided the recombination processes are negligible, otherwise it is necessary to introduce one more term K_2N^2). This relationship represents a standard curve with saturation, like the curve 1 in Fig. 1. However, other experimental data presented above point to the fact that the situation can be much more complicated if the accumulation curve becomes non-monotonic, that is related to simultaneous formation of another type of defects.

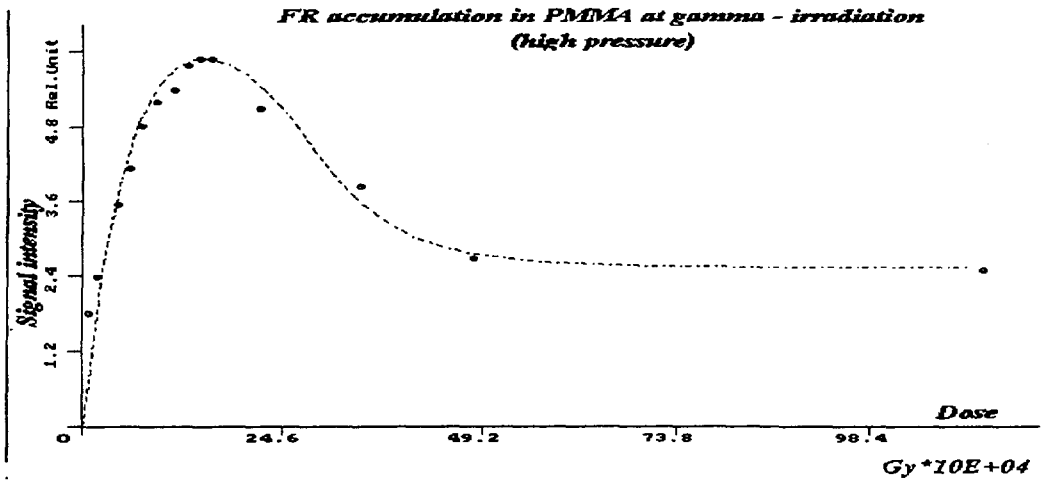
The mechanism of "secondary" radical production can be caused by the heat of recombination of the initial radicals as a result of the radiation field influence. The recombination energy release causes local heating in the matrix and changes it's composition, therefore "secondary" radicals can appear there.

These processes can be taken into account by the set of two equations for the "primary" defects N_1 and the "secondary" ones N_2 [5]:

$$\begin{cases} \dfrac{dN_1}{dD} = 1 - C_1 N_1 N_2 - C_2 N_1 \\ \dfrac{dN_2}{dD} = C_3 N_1 N_2 - C_4 N_2 + C_5 \end{cases} \tag{3}$$

Here the factors C_2 and C_4 are similar to the constants K_1 in Eq. (2), C_1 and C_3 take into account the formation of the N_2 defects at the expense of the annihilation of the N_1 defects and C_5 is related to the mechanism of direct generation of the defects N_2 by radiation.

a)



b)

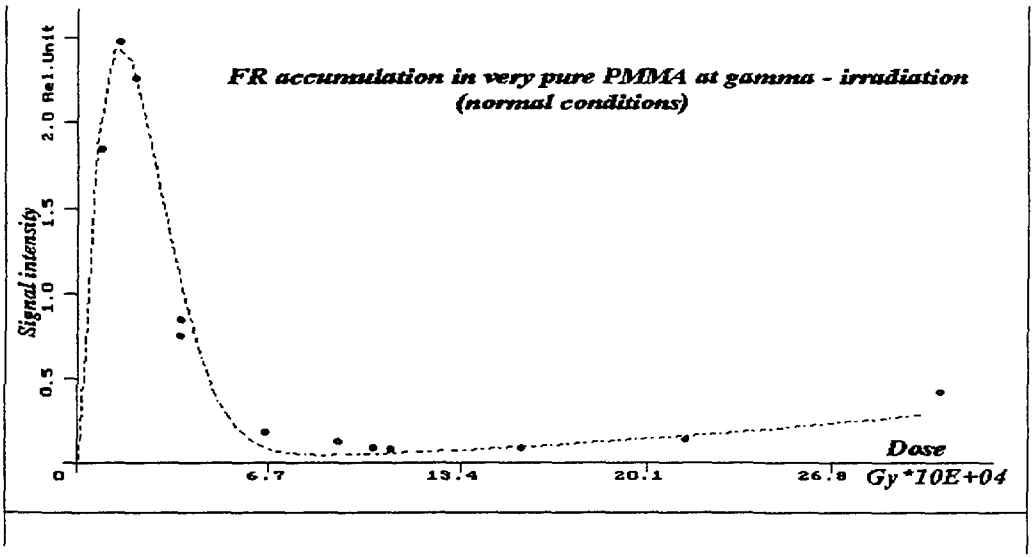


FIG. 5. Fitting to the experimental accumulation curve in the frame of the models: points represent experimental data, line represents the model; a - model (3), and b - model (4)

The other approach, close to this one, is related to the introduction of a "damaged volume" concept. According to this model, the primary defects can be produced only in an initial intact matrix. As a result of its interaction with the radiation field, however, they disappear in a matrix and a "damaged volume" is formed locally, i.e. it's structure changes. The secondary defects can be produced only within this "damaged volume", thus they can also interact later with the radiation field. Occurrence of the "damaged volume " is determined simply by the Poisson's distribution:

$$\begin{cases} \dfrac{dN_1}{dD} = \exp(-C_1 D) - C_2 N_1 D \\ \dfrac{dN_2}{dD} = \left[1 - \exp(-C_3 D)\right] - C_4 N_2 D \end{cases} \qquad (4)$$

The conformity of the models described by Eqs (3) and (4) to the experimental data is illustrated by Figs 5a and 5b.

3. CONCLUSION

The data presented here (its number could be increased) show that it seems to be reasonable, in view of ESR-dosimetry, to separate, at least, three stages of radiation damage in solids:

- the initial stage (small doses), when the defect concentration is low, they, practically, don't interact between each other, and the relationship of defects accumulation versus the dose is close to linear;
- the intermediate stage, when interaction between defects occurs, as a result, the relationship of accumulation noticeably differs from the linear one, and a non-monotonic character can occur;
- the high-dose stage, when the significant damaged volume has been accumulated (paramagnetic defects can be even annealed and, from the point of view of ESR, the sample may be represented as unirradiated or as irradiated by a small dose) and all peculiarities of the second stage manifest itself more prominently.

For various substances and various conditions of irradiation, the effects of the second and the third stages manifest themselves at different values of irradiation dose. For example, for PMMA the second stage is obviously observed already at 5 kGy as the irradiation temperature increases, and in α-alanine the relationship of accumulation is close to linear up to 100 kGy; nevertheless, the same effects occur.

Apparently, the marked peculiarities are characteristic of all solids, e.g., in Refs [5, 6] data about "radiation memory" of metals and alloys even after deep annealing up to the melting condition are presented.

Thus, for correct application of ESR- dosimetry methods it is necessary either to ensure the conditions of the first stage, or to take into account potential nonlinearities arising, for example, on the basis of the model like (3) or (4); otherwise very significant errors can appear.

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