Human EPR Dosimetry at Low Accumulated Dose of Ionizing Radiation

Valeri E. Galtsev, E.V. Galtseva, Ya.S. Lebedev, Moscow

Abstract

A new method for tooth (enamel and dentine) and bone EPR dosimetry is proposed that enhances substantially the dosimetric sensitivity. The method is based on the nonstandard spectra registration mode (the second harmonic phase quadrature detection of absorption under rapid passage conditions). This method provides an apparent enhancement of the component ratio of radiation to a nonradiative background (approximately fivefold) and allows to increase the radiation signal to noise ratio (approx. twofold) in comparison to the standard method used in EPR dosimetry. The data handling procedure is more simple and reliable than the standard mode procedure.

Introduction

A wide exploitation of ionizing radiation sources in medicine and industry, and the possibility of occuring accidents at nuclear power stations led to the development of dosimetric methods. Tooth Electron Paramagnetic Resonance (EPR) dosimetry became one of the most attractive methods either for human and animal retrospective or for accident dosimetry [1, 6, 10]. Tooth EPR dosimetry is of a significant practical interest due to the possibility of using human and animal teeth as individual dosimeters. The retrospective dosimetry is of highest importance for the dose evaluations of inhabitants of regions with radioactively contaminated soils. In such cases any available data concerning the extent of the radiation effect on inhabitants are necessary to be determined if urgent medical treatment and any activity aimed at elimination of the pathogenic irradiation effect is needed.

The EPR method was applied for a long time as a tool to study radiation effects. Under ionizing irradiation unpaired electrons in materials are produced, whose concentration is dependent on the absorbed dose. Therefore the latter can be determined from the EPR spectrum of the sample. Radiation induced center' lifetime in solid specimens (minerals, teeth, bones, mollusk shells, ets.) is long enough (ca. 10^9 years for tooth enamel, for example [14]) to be detected. This procedure allows to determine, with a high accuracy, the total dose absorbed by the sample by using the signal intensity of radiation induced unpaired electrons. The following advantages are pertinent to EPR in comparison to other dosimetry methods (e.g. thermoluminescence, or spectrophotometry):

1) the possibility of nondestructive analysis;
2) quantitative dose evaluation;
3) high sensitivity and large dynamic range of dose evaluation.

The tooth EPR dosimetry procedure is as follows: The samples are extracted human tooth pieces from which any dentine-and caries-damaged parts are carefully removed. For these prepared samples, the EPR signal is measured either before or after a series of artificial irradiation (in steps of 40-200 cGy 4-5 irradiations in all). The dependence of the EPR signal on the artificial irradiation dose is extrapolated to the zero value of the EPR signal and an appropriate dose value is ascribed to the dose accumulated by the sample (initial dose Do at the investigation momentum).

Despite the great advances in human tooth EPR dosimetry, there are some not yet solved problems which diminishes the reliability of the results, and restricts the practical applicability of the method. One of the problems is insufficient sensitivity of the method. The sensitivity of the method is restricted by the presence of the nonradiation background component that overlaps with the radiation one. This is crucial for a vital dose range (10 - 200 cGy) for human dosimetry (Fig.1A). The proposed methods of chemical treatment of tooth enamel and bone tissues make it possible to decrease, to some extent, the background components (approx. twofold for tooth enamel [13], and fivefold for dentine and bone [17]), but they are not able to eliminate it totally. The standard method of EPR dosimetry [1,6,10] is based on the registration of the first derivative signal of absorption (in phase with the magnetic field modulation - Standard Detection Mode - SDM) at room temperature (Fig.1A). The application of this procedure requires the extraction of the radiation component from the experimental spectrum for which the following methods were proposed:

1) The subtraction of the background component:

a) computer simulated (for example: approximated by the Lorentzian line with parameters selected to fit the low-field part of the EPR signal, the latter supposed to be almost free from overlapping with radiation component [7]);
b) approximated by a signal of nonirradiated milk-tooth enamel [15].

The whole procedure for such methods becomes complicated and involves many sources of errors at low initial doses when the radiation component looks like a hardly observable perturbation of the background.

2) The selective use of saturation which is based on the difference in the microwave power dependences of the radiation component of EPR spectrum and the background one. The subtraction of the spectrum recorded at low microwave power from that recorded at high microwave power provides a reduction of the background component in the spectrum [8,12,16]. However, the shape of the background component (for tooth enamel) depends on the microwave power. So, the selective saturation is proven to be inapplicable to EPR dosimetry.

Therefore, there is a practical interest in finding some new ways which provide a more effective discrimination of the radiation component from background. The starting assumption for this research was the fact that these components possess different relaxation parameters. In the present work, a new method of EPR dosimetry is proposed. This method is based on using the (for EPR dosimetry) nonstandard registration mode (second harmonic phase quadrature detection of absorption at rapid passage conditions - Rapid Passage Mode). This Rapid Passage Mode (RPM) is rather sensitive to relaxation characteristics of the spin system and helps to permit, in some cases, to extract signals from a complicated spectrum [2, 5]. This detection mode is available at most of the commercial EPR spectrometers. Here, the EPR dosimetry procedure is described based on the RPM and the comparison is made for low accumulated dose measurements in RPM and in SDM.

Method and Materials

Spectra were recorded with a commercial X-band Bruker EPR spectrometer ESP-300, under two registration conditions:

1) Standard Detection Mode (SDM - first derivative of absorption) at room temperature, and

2) Rapid Passage Mode (RPM - second harmonic phase quadrature detection of absorption) at 77K.

The modulation frequency was 100 kHz, and the modulation amplitude 0.32 mT. The microwave power was 3 - 5 mW. The rectangular microwave cavity of TE_{102} mode was used.

Teeth were obtained from patients in the normal course of dental practice. Tooth enamel samples (milk tooth - sample I, adult tooth - sample II) for EPR measurements were made by removal of the dentine and caries-damaged parts of enamel. A dentine sample (sample III) was made by removal of the enamel and caries-damaged parts of dentine.

The samples were prepared as cylinders of 5 mm diameter and 5 mm height of small pieces of

enamel or dentine (~ 0.3 mm) embedded in a polystyrene matrix. Special experiments proved that polystyrene, both irradiated or nonirradiated, did not produce any noticeable EPR signal. The mass of enamel (or dentine) in each sample was about 100 mg.

Special attention was paid to the quality of the Dewar vessel as the quartz defects signal can mask the signal of interest when RPM at 77K is applied.

The sample irradiation was performed with a standard ^{60}Co γ-ray source. The dose value applied was monitored with thermoluminescent detectors. For complete annealing of short-lived radiation radicals, the samples were kept during 4 h at 60°C.

Experimental results and discussion

As a result of our investigations the optimal spectra registration conditions in RPM were determined from the point of view of the radiation-to-background component ratio and signal-to-noise ratio [3,4]. In the present work, the dosimetry procedure is proposed based on the rapid passage mode, and a comparison is carried out between the application to the accumulated low dose measurement of RPM and SDM.

Tooth enamel.

Fig. 1A shows the EPR spectra of sample I (milk tooth enamel) recorded in SDM after the exposure to several irradiation doses. It can be easily realized that in all the spectra, the radiation component is considerably masked by the background one. With the additional radiation dose of 2.0 Gy, the ratio of the amplitude of the radiation component ($I^{max}rad$ obtained by subtraction the background component using standard procedure [17]) to the background one ($I^{max}bg$) equals:

$$I^{max}rad / I^{max}bg \approx 0.8.$$

Fig. 1B represents the spectra of the sample I recorded in RPM. It can be deduced that the background component masks the radiation component considerably less, and, in this mode, a much more favorable ratio $I^{max}rad/I^{max}bg \approx 5.5$ (for the same additional radiation dose 2.0 Gy) is achieved. Thus, in RPM in comparison to SDM, the ratio of the amplitude of the useful radiation component in relation to the background one increases substantially.

Fig. 2 shows the dependence of the radiation component's amplitude on the irradiation dose, achieved by the analysis of the recorded RPM spectra of sample I. The data handling procedure is as follows. For the signal intensity at the positions "1" and "2" of the experimental spectrum we have

$$I_{exp}^{(1)}(D) = I_{rad}^{(1)}(D) + I_{bg}^{(1)}, \quad (1)$$

$$I_{exp}^{(2)}(D) = I_{rad}^{(2)}(D) + I_{bg}^{(2)}, \quad (2)$$

where "1" corresponds to the position of the radiation component maximum, "2" is the point symmetrical to position "1" with respect to the background component center. With a reasonable accuracy, the background component's shape in RPM can be

approximated by a symmetrical line (no matter of which shape), so

$$I_{bg}^{(1)} = I_{bg}^{(2)}, \qquad (3)$$

As

$$I_{rad}^{(2)} = \alpha I_{rad}^{(1)}, \qquad (4)$$

from (1), (2), and (3) we obtain

$$I_{rad}^{(1)}(D) = \left\{ I_{exp}^{(1)}(D) - I_{exp}^{(2)}(D) \right\} / (1-\alpha) \qquad (5)$$

The shape of the radiation component is independent of the radiation dose, ad minimum for doses less than 10 kGy, which is much higher than doses of practical human dosimetry interest. The value of α can, hence, to a sufficient accuracy, be determined from the spectrum of the sample exposed to a high dose. In our case $\alpha \approx 0.045$ (D = 20 Gy). This minimum value of α suggests to a high degree of precision

$$I_{rad}^{(1)}(D) = I_{exp}^{(1)}(D) - I_{exp}^{(2)}(D = 0) \qquad (6)$$

where $I_{exp}^{(2)}(D = 0)$ is the experimental spectrum intensity at the position "2" without additional irradiation dose. The values of initial irradiation dose obtained from the dose dependences "o" (5), "+" (6) are close to each other and equal $D_0 \approx 4$ cGy.

Fig. 2 also shows the dependence of the amplitude of the radiation component on the irradiation dose, achieved by the same analysis of the recorded RPM spectra of sample II. The values of initial irradiation dose obtained from the dose dependences "•" (5), "Δ" (6) are close to each other and equal $D_0 \approx 23$ cGy.

Dentine.

The difficulties of determining the accumulated radiation doses in dentine and bone tissues are the same as that for tooth enamel, namely, the strong masking of the useful radiation component by the background component [9, 11].

Fig. 3 shows the EPR of sample III (dentine) exposed to various doses of artificial radiation, recorded in SDM (A) and in RPM (B). It can be realized that, in the latter case, the radiation component is masked by the background component to a considerably lower extent than in the former case (SDM). Thus, with an additional radiation dose of 16 Gy the radiation-to-background component ratio for the spectrum recorded in SDM is about $I^{max}rad/I^{max}bg \approx 0.8$, while for the spectrum recorded in RPM the same ratio is about $I^{max}rad/I^{max}bg \approx 4$.

Hence, the EPR dosimetry method based on registration of spectrum in RPM proposed in this work, makes it possible

i) to obtain a much more favorable ratio of the useful amplitude of the radiation component to the background one that provides an increase of the sensitivity of the EPR dosimetry;

ii) to use more reliable and simple experimental data handling procedure which, in comparison to the standard method [6]; results in a decrease of errors of the dose evaluation at low accumulated doses

iii) to increase the radiation signal-to-noise ratio about twofold which then results in the advantage that one can decrease the sample mass necessary for EPR dosimetry (or to diminish the spectrum registration time).

References

1. Aldrich J.E. and Pass B. Determining Radiation Exposure from Nuclear Accidents and Atomic Tests using Dental Enamel. Health Phys. 54(4), 469 (1988).
2. Feher G. Electron Spin resonance Experiments on Donors in Silicon. Phys.Rev. 114, 1219 (1959).
3. Galtsev V.E., Grinberg O.Ya., Lebedev Ya.S. and Galtseva E.V. EPR Dosimetry sensitivity Enhancement by Detection of Rapid Passage Signal of the Tooth Enamel at Low Temperature. Appl.Magn.Res. 4, 331 (1993).
4. Galtsev V.E., Galtseva E.V. and Lebedev Ya.S. Optimal Registration Conditions for Tooth EPR Dosimetry at Low Accumulated Dose. Book of Abstracts 4th International Symposium on ESR Dosimetry and Applications, Munich, May 1995.
5. Hyde J. NMR and ESR-spectroscopy, Moscow (in Russian) 1964.
6. Ikeya M. New application of Electron spin resonance. Dating, Dosimetry and Microscopy, Singapore, (1993).
7. Ishii H., Ikeya M. and Okano M. ESR Dosimetry of Teeth of Residents Close to Chernobyl Reactor Accident. J.Nucl.Sci.Tech., 27(12), 1153 (1990).
8. Moroz I.A., Serezhenkov V.A., Klevezal G.A., Burbaev D.S. and Vanin A.F. New opportunities in EPR Dosimetry. Biophysics, 39, 1075 (in Russian) (1994).
9. Ostrowski K., Goctawska A.D., Stachwicz W. Stable radiation-induced paramagnetic entities in tissue mineral and their use in calcified tissue research. Free Radicals in Biology (Ed. Pryor W.A.), 4, 321 (1980).
10. Pass B. and Aldrich J.E. Dental enamel as an in vivo dosimeter. Med.Phys. 12(3), 305 (1985).
11. Pass B., Aldrich J.E. and Scallion P.L. An Analysis of Paramagnetic Centers in Irradiated dentine Using Electron Spin Resonance. Calcif. Tissue Int. 46(3), 166 (1990).
12. Romanyukha A.A., Regulla D., Vasilenko E. and Wieser A. South Ural Nuclear Workers: Comparison of Individual Doses From Retrospective EPR Dosimetry and Operational Personal Monitoring. App. Radiat. Isot. (in press), (1995).
13. Schwartcz H.P. Nucl.Tracks, 10, 865 (1985).
14. Serezhenkov V.A., Domracheva E.V., Klevezal G.A., Kulikov S.M., Kuznetsov S.A., Mordvintcev P.I., Sukhovskaya L.I., Schklovsky-Kordi N.E., Vanin A.F., Voevodskaya N.V. and Vorobiev, A.I. Radiation Dosimetry for residents of the Chernobyl Region: A Comparison of Cytogenetic and Electron Spin Resonance Methods. Radiat.Prot.Dosim. 42(1), 33 (1992).
15. Serezhenkov V.A., Moroz I.A., Klevezal G.A. and Vanin A.F. Estimation of Accumalated Dose of Radiation by the Method of ESR-Spectrometry of Dental Enamel of Mammals. Book of Abstracts 4th International Symposium on ESR Dosimetry and Applications, Munich, May 1995.
16. Romanyukha A.A., Ignatiev E.A, Koshta A.A. and Wieser A. Selective saturation method for EPR dosimetry with tooth enamel. Book of Abstracts 4th International Symposium on ESR Dosimetry and Applications, Munich, May 1995.
17. Wieser A., Haskell E., Kenner G. and Bruenger F. (1994) EPR Dosimetry of Bone Grains Accuracy by Isolation of Calcified Tissue. Appl.Radiat.Isot. 45(4), 525 (1994).

Fig.1.
EPR spectra of sample I (milk tooth enamel) at several additional γ-radiation doses as detected in SDM (A) and in RPM (B). The spectrum of the sample exposed to 20 Gy either in SDM or RPM was recorded with 3,3 -fold attenuation.

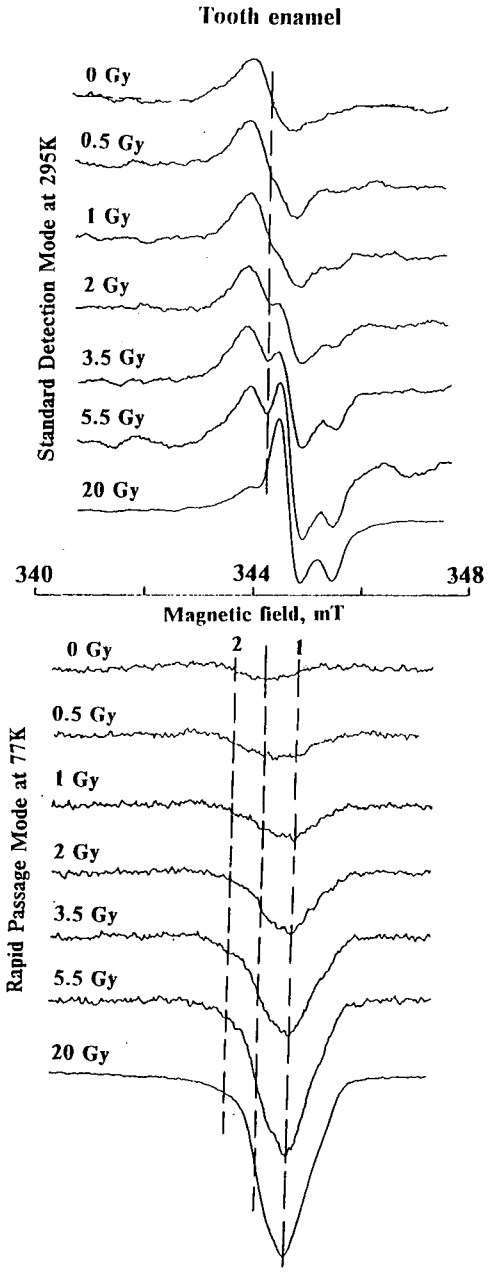

Fig.2.
The radiation component amplitude in RPM versus additional γ-radiation dose:

1) sample I	**o derived by (5),**	**+ derived by (6);**
2) sample II	**• derived by (5),**	**Δ derived by (6).**

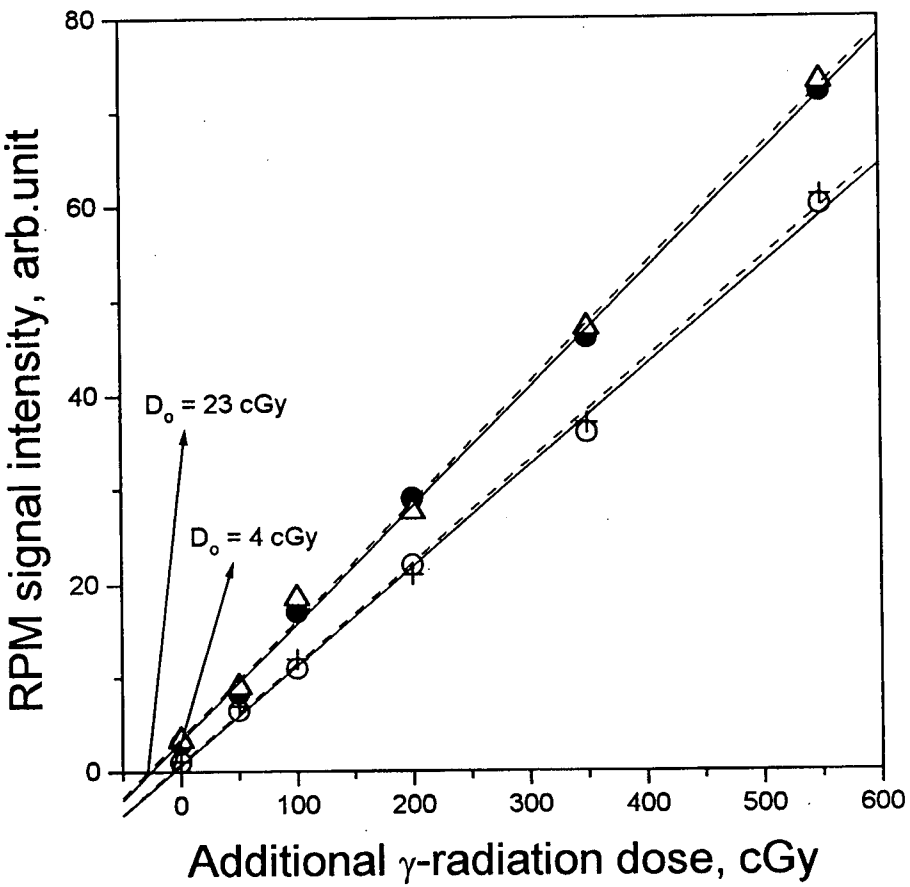

Fig.3.
EPR spectra of sample III (dentine) at several additional γ-radiation dose as detected in SDM and in RPM.