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EPR DOSIMETRY BY TOOTH ENAMEL: UNCERTAINTIES AT DOSE RECONSTRUCTION

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The retrospective EPR estimation of individual irradiation dose is based on the use of human tooth enamel as a radiation detector. Ionizing radiation creates stable CO_3^{3-} radicals in such enamel, which later can be determined measuring the amplitude of EPR signal proportional to the number of inserted radicals and to the absorbed dose. The method was applied to such categories of people as survivors of the atomic bomb explosion in Japan, nuclear workers in South Ural, and liquidators of the consequences of the Chernobyl reactor's accident. The results show that the EPR method is applicable for dose reconstruction down to the 30 mGy level, with special precautions to be taken at such a low-level exposure. The paper is focused on the estimation and analysis of the EPR measurement uncertainties at dose reconstruction by tooth enamel. The authors also analyze the contribution of uncertainties connected with equipment, sample preparation, radiation sensitivity calibration, signal processing, *etc.*, to the total standard uncertainty of the EPR method.

Keywords: *EPR dosimetry, measurement uncertainty, dose reconstruction, tooth enamel.*

1. INTRODUCTION

The method of EPR spectroscopy of human tooth enamel (the EPR dosimetry) is becoming internationally recognized as the technique for determination of individual accumulated doses in accidental [1, 2] and retrospective [3, 4] dosimetry.

The method of retrospective EPR dosimetry using tooth enamel is based on measuring the number of radiation-induced radicals in hydroxyapatite (HAP) $Ca_{10}(PO_4)_6(OH)_2$ – a mineral component of teeth and bones. The EPR technique requires only a simple sample preparation, giving a precision of about 30 mGy (1 sigma) for a deciduous incisor [5].

In previous studies [6], the individual doses were estimated by the EPR method for a number of Latvian specialists that had dealt with liquidation of the

consequences of Chernobyl reactor's accident. The retrospective estimation indicated individual doses in the range 100–500 mGy – higher than those officially documented. However, this estimation included some uncertainties related to the medical exposure, environmental background contribution, metabolic modification, *etc.*

Additional uncertainties in EPR dosimetry could arise as the result of non-perfect sample preparation, scatter in irradiation conditions, instability of spectrometer operation, and so on. The purpose of this paper was to find and estimate the sources of errors (uncertainties) at the EPR retrospective dosimetry by tooth enamel.

2. MATERIALS AND METHODS

To estimate possible uncertainties at the EPR dose reconstruction by tooth enamel, a concept similar to that presented in the ISO recommendations [7] was used. According to the recommendations, uncertainty of a measurement is the difference between the result of the measurement and the true value. According to [7], type A and type B uncertainties are distinguished: those evaluated by statistical and non-statistical methods, respectively.

The present work employs an approach similar to that recommended by [8] for uncertainty evaluation. The following uncertainties could be related to the measurement of an individual dose absorbed by tooth enamel:

- sample preparation uncertainty (U_{sam});
- fading uncertainty (U_{fad});
- uncertainty of measurement (U_{EPR});
- uncertainty of metabolic modification (U_{met});
- uncertainty of natural background dose (U_{back});
- uncertainty of dose response calibration (U_{cal}).

The listed uncertainties could be grouped based on the assumption that there is no correlation between the components of the following expression for the total uncertainty:

$$U_{EPRDOSE} = \left(U_{sam}^2 + U_{fad}^2 + U_{EPR}^2 + U_{met}^2 + U_{back}^2 + U_{cal}^2\right)^{1/2}. \quad (1)$$

In the next section the $U_{EPRDOSE}$ components are discussed.

3. RESULTS

Sample preparation uncertainty

Sample preparation technology is the crucial part of dosimetry [9]. The technology used in the given studies consisted of the steps shown in Fig. 1.

Enamel was carefully separated from the dentine by a hard-alloy dental drill minimizing the influence of dentine impurities on the EPR signal from enamel (care was taken not to induce additional mechanical signals that would interfere with the radiation-induced signal [10]). The enamel was crushed by an agate mortar to small chips with a linear size of 0.1–1 mm.

One of the technological steps was weighting the enamel powder for measurements. The crushed enamel powder was weighted by a SATORIUS BL105S analytical balance with the accuracy of 1 mg. To achieve good repeatability of EPR

spectra, the average weight of the sample was kept 100 mg. The weighting uncertainty U_w was 1%, which is negligible compared with the total uncertainty of sample preparation.

The enamel samples were washed in ethanol and dried in annealed silica gel to decrease the spectral noise caused by impurities from water.

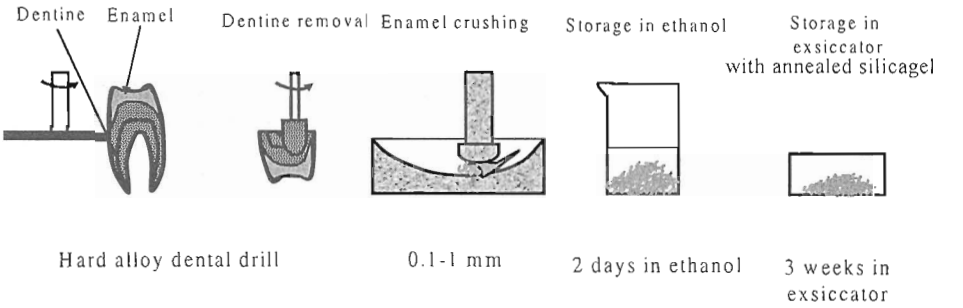

Fig. 1. Sample preparation technology.

The sample preparation technology was verified by the EPR sensitivity measurements on ten non-irradiated samples. The average uncertainty of the sample preparation technology was found to be ~8% (the type B uncertainty reflected in the sample radiation sensitivity).

For the calculations the total uncertainty of sample preparation was taken 10%.

Fading uncertainty

Fading or decreasing of the dosimetric signal is a common problem in many dosimetry techniques, such as film technique, thermoluminescence, EPR, *etc.* Fading was observed for the EPR signals created by mechanical treatment of the sample [10], exposure to sunlight [11], metabolical modification [12], caries, and so on. However, according to the palaeontological dating EPR studies [8], the lifetime of CO_2-radicals in enamel was estimated to be 10^7 years for fossil enamel. Indications exist that the radiation-induced CO_2-radicals in the tooth enamel used for dose reconstruction are stable *in vivo*, i.e. during a human lifetime [8]. Thus, there is no evidence that the fading contributes to the total uncertainty of EPR dose reconstruction by tooth enamel. Taking this into account, we can assume $U_{fad} = 0$ (type B uncertainty).

EPR measurements

The EPR measurement uncertainty (U_{EPR}) is a combination of uncertainties from spectrometer noise (U_{noise}), sample positioning (U_{posit}) and spectrometer stability (U_{stab}):

$$U_{EPR} = \left(U_{noise}^2 + U_{pos}^2 + U_{stab}^2\right)^{1/2}. \qquad (2)$$

These factors are instrument-related, being highly dependent on the spectrometer design and parameters.

EPR spectrum noise

The signal intensity at the EPR dosimetry of tooth enamel is overlapped by the noise in the spectrum from high-frequency and low-frequency sources. The high-frequency noise is electronic in character and can be taken into account by averaging the spectra. In the given measurements this was done for 20 to 30 spectra. The low-frequency noise appears in the spectrum as fluctuations whose line width is comparable with that of EPR signal. Both these characteristics are highly dependent on the performance of the EPR spectrometer, being equal to 10–12% for the given setup (PE-1306, Minsk, USSR). For future calculations a spectral noise level U_{noise} = 12% was assumed for the given amplitudes of analyzed signals (type A uncertainty).

Sample positioning

The EPR signal intensity is influenced by variations (angular and vertical) in the sample positioning in a microwave cavity. The variations are usually caused by tolerances in the sample tube dimensions, non-uniform mass distribution of the sample, and the EPR signal anisotropy. The reproducibility of sample positioning in our measurements was improved by running three measurement sessions after rotating the enamel powder samples. Type A uncertainty, U_{po}, was determined experimentally and found to be ~2%.

Spectrometer stability

The stable operation of a spectrometer is directly associated with its sensitivity as one of the most important parameters affecting the accuracy of EPR measurements. This becomes especially important in the case of using such dated EPR spectrometers as PE 1306, with the sensitivity variations up to 10% and even greater.

The stability of an EPR spectrometer is determined by the microwave power output, the cavity quality factor, and the applied magnetic fields. These parameters are influenced by the temperature and humidity of the sample and equipment. In the given case, the spectrometer stability was verified by a MgO (Cr^{3+}) standard crystal inserted in the resonator together with the sample. This allowed the uncertainty of spectrometer stability, U_{stab}, to be decreased down to ~ 7%.

The total uncertainty of EPR measurements, U_{EPR}, including U_{noise}, U_{pos}, U_{stab}, was 14% for the given amplitude of EPR signal (a dose below 1 Gy).

Contribution of natural background radiation

The contribution of the natural background radiation accumulated during a human's lifetime between the tooth formation and the measurements should be subtracted from the total dose measured. This dose for Latvian inhabitants is typically 1 mGy per year (the value was obtained using TLD dosimeters and calculated assuming the average terrestrial exposure to be 100 nGy/hour).

The age of a tooth differs from that of a person, depending on its location and therefore on the time of its formation [13]. The biggest difference is observed for children teeth, where contribution of the given factor is substantial. For adults this difference is observable only for the so-called wisdom teeth, which are formed only at the age of 20–25 years. In the given research no wisdom teeth were studied,

therefore the age of teeth was taken equal to a person's age. Deviations from this assumption produce the uncertainty of dose reconstruction, U_{back}, about 3%.

Metabolic modification of enamel

A discussable matter is the contribution of tooth enamel metabolism to the EPR signal and its dependence on the individual. According to [14], such metabolism can seriously modify the EPR spectra and the fading dosimetric line. The authors doubt about the EPR dosimetry accuracy, arguing that there should be high contribution of metabolic processes on enamel. However, in the opinion of other authors [2, 3, 8], any noticeable metabolism is hardly possible since tooth enamel is the most resistant calcified tissue in a human organism. In our studies its contribution to the total uncertainty was accepted 10% and treated as type A uncertainty. This value has been obtained for no caries-affected teeth and is based on the personal experience while working with calcified tissues. Therefore, for further calculations U_{met} = 10% could be taken.

Dose calculation/reconstruction method

For measuring the dose signal two approaches exist. The first one is individual calibration of the enamel sensitivity for each tooth sample – the so-called additive dose method. The second uses the average sensitivity of the enamel to radiation and is called the spectrum subtraction method.

According to the first method it is necessary to obtain the dependence of the radiation-induced signal on the irradiation dose. This method is based on the saturation of the background signal due to irradiation and a linear growth of the dosimetric signal. The sample is cumulatively exposed to irradiation with corresponding measurements of the amplitude of EPR dosimetric signal. The initial absorbed dose is derived from the intercept of the linear regression line with the dose axis. Figure 2 shows the linear dependence and the least squares fitting.

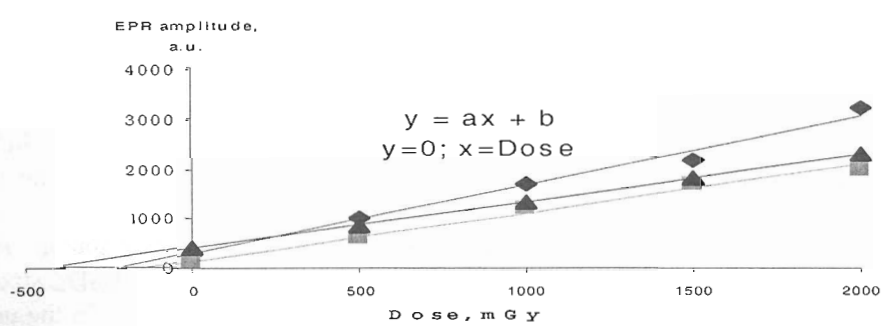

Fig. 2. Initial dose reconstruction by the additive dose method.

The second method – that of spectrum subtraction – is distinguished by the procedure used for elimination of the native background signal and measurement of the dosimetric signal intensity [14]. The method of background spectrum substraction for dose reconstruction is non-destructive and much less time-

consuming. It is based on the assumption that the variations in radiation sensitivity in the enamel of different individuals are only moderate.

The ESR spectrum of a non-irradiated reference sample is subtracted from the spectrum of the investigated sample. The reference sample prepared of homogenized enamel material taken from several adults is selected in such a way that it has the most symmetric EPR line. The amplitude of the dosimetric signal is measured in the resulting difference spectrum (Fig. 3).

The total uncertainty is calculated using the curve fitting algorithm and individual sensitivity variation. For instance, applying as such algorithm a Gaussian-type curve one can obtain the uncertainty of approximation close to that of EPR signal measurement (~14%). However, the experimentally observed radiation sensitivity of the tooth enamel strongly depends on the individual. Variations in the radiation sensitivity for different individuals can reach 80–100% of the difference for the samples. Therefore, to reduce the total measurement uncertainty the additive dose method is preferable for EPR dose reconstruction.

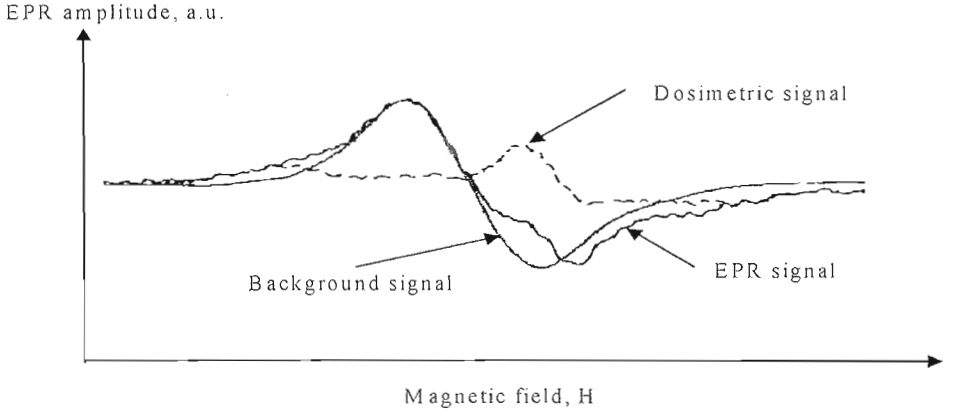

Fig. 3. Spectrum subtraction method illustrated by a typical spectrum of tooth enamel.

Uncertainty of additional irradiation

According to the additive dose method used in the given studies, the irradiation dose dependence of the radiation-induced signal is to be obtained. In the experiment, a linear dependence was assumed and the least squares fitting was used.

The tooth enamel samples were irradiated at the Secondary Standard Dosimetry Laboratory (SSDL) of the Latvian Metrological Center. In the SSDL studies the combined (type A + type B) uncertainty at the dose determination in the units of air kerma was about 2%. This value held true for a fixed distance only (usually 1 m), fixed radiation field sizes, and well-controlled irradiation time.

A Cs-137 source with a well-defined output was selected for enamel irradiation. The dose absorbed by the tooth enamel was estimated by the formula:

$$D = \dot{D}_k \times t \times k\,, \tag{3}$$

where D_k is the air kerma dose rate, t is the time of irradiation, k is a conversion factor of the expression for absorbed dose in HAP [15].

Uncertainties of irradiation could be estimated based on the following assumptions.

a) The uncertainty of the air kerma dose rate is the sum of positioning uncertainty U_{pos} and combined source uncertainty U_{source}.

b) The tooth enamel was irradiated at a 1 m distance from the source with the positioning accuracy of 1 cm (distance uncertainty $U_d = 1\%$). The positioning uncertainty U_{pos} was estimated by the inverse square law of radiation propagation, and for the given setup it was 2 % (type A uncertainty).

The combined uncertainty of source U_{source} includes additional factors that are influencial as to the dose delivered to the enamel – such as variations in the air temperature, humidity, pressure, and spontaneous disintegration of the source. For the given setup the contribution of all these factors was about 4 % (type B uncertainty).

The irradiation time was controlled by a laboratory clock indicator (accuracy 1 s). For a 1 Gy dose it was 200 min. The uncertainty of irradiation time determination U_t was below 1 %.

The correction factor k was estimated based on the Monte-Carlo simulation of the electron transport in tooth enamel. The value k was taken from [16] and the uncertainty was assumed to be below 3%.

The uncertainty of EPR dose calculation/reconstruction includes all the above mentioned uncertainties. Assuming that these do not correlate with each other, one can obtain:

$$U_{cal} = \left(U_p^2 + U_t^2 + U_{source}^2\right)^{1/2}. \qquad (4)$$

The uncertainty of EPR dose calculation, U_{cal}, was estimated equal to 0.055 (for the future studies 6% were accepted).

4. DISCUSSION

Estimation of the total uncertainty of EPR dose reconstruction

Based on the estimations given above for different contributions to the total uncertainty of EPR dose reconstruction, the $U_{EPRDOSE}$ value was found to be <20% for the dose interval 100–500 mGy (including the mentioned contributions: U_{sam}, U_{EPR}, U_{mel}, U_{back}, and U_{cal}). This uncertainty should be multiplied by a coverage factor of 2.0 to obtain the overall combined uncertainties at the 95% confidence level. This uncertainty is only valid for photon exposures with the energies above 300 keV.

The contribution of U_{fad} was neglected in the analysis of uncertainties due to its relatively weak influence on the total uncertainty.

The given estimation of combined uncertainty could be characterized within the limits of an ideal case not including some important contributing factors. When reconstruction of the personal dose accumulated by a human is concerned, other sources of uncertainty often making a major contribution to the total value may be involved. A number of these uncertainties and their values depend on a particular application. The sources of these uncertainties are well known, however quanti-

fication and reduction of their contribution are often problematic. The former needs special consideration in each particular case and is beyond the scope of this paper.

Typical sources of uncertainties associated with reconstruction of the personal dose are listed below.

- Unknown photon energy spectra for the radiation fields.
- Mixed field irradiation by different sources of alpha, beta, gamma and neutron irradiation.
- Lifetime irradiation of people at medical treatment, particularly when low energy photons are involved.
- Solar UV exposure on the rear part of incisors and canines, the cases of partial body exposure.
- Unknown irradiation geometry.
- Tooth enamel metabolism (according to [14] seriously contributing to the EPR signal).
- Tooth diseases and other individual specific parameters.

5. CONCLUSION

Based on the estimations given above the total uncertainty of EPR dose reconstruction is ~ 20% (1 sigma interval) for the doses below 500 mGy. This value could be assumed as a first approximation that does not take into account such individual specific factors as exposure history and metabolic modification of tooth enamel.

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EPR DOZIMETRIJA, IZMANTOJOT ZOBU EMALJU: DOZAS REKONSTRUKCIJAS NENOTEIKTĪBA

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Kopsavilkums

Ar elektronu paramagnētiskās rezonanses (EPR) metodi izmērīto individuālo dozu retrospektīvā novērtēšanā cilvēku zobu emalju izmanto kā radiācijas detektoru. Jonizējošais starojums emaljā rada stabilus CO_3^{3-}radikālus, kurus vēlāk var izmērīt ar EPR. EPR signāla amplitūda ir proporcionāla radīto radikālu koncentrācijai un atbilstoši absorbētai dozai. EPR metode izmantota, lai novērtētu individuālās apstarošanas dozas Japānā atomu bumbu sprādzienos cietušajiem, kodolenerģētikā strādājošiem Dienvidurālos, Krievijā, Černobiļas AES avārijas "likvidatoriem". Šo pētījumu rezultāti apliecina iespēju izmantot EPR metodi dozu rekonstrukcijai no 30 mGy. Tomēr, lai novērtētu tik zemu apstarošanas līmeni, jāievēro speciāli priekšnoteikumi. Raksts veltīts iespējamo nenoteiktības avotu izvērtēšanai un analīzei zobu emaljas dozu rekonstrukcijā ar EPR metodi. Analizēti dažādu faktoru (mēriekārta, paraugu gatavošana u.c.) ieguldījums EPR metodes standarta nenoteiktībā

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