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APPLICATION OF THE OPTIMIZATION THE PARAMETERS OF MEASURING TOOTH ENAMEL EPR SPECTRA FOR EMERGENCY HUMAN DOSIMETRY

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It is known from the world literature that for EPR spectrometry for emergency dosimetry, where doses from 0.5 Gy and higher are used, the quality and reliability of calculating the experimental dose depends on the parameters of recording spectra (accumulation time, microwave power).
This article describes the results of the development and application of a method for optimizing the parameter of recording EPR spectra on human tooth enamel, such as microwave power, but in the range of emergency radiation doses (from 1 Gy to 5 Gy), under conditions of a local radiation incident based on a biodosimetric laboratory. The results showed that the obtained experimental microwave power value of 5 MW is the most effective parameter for improving the quality of EPR spectra of human tooth enamel for a range of emergency radiation doses, even taking into account different types of spectrometers on which measurements are carried out. Optimization of the parameter for recording EPR spectra, in particular microwave power, improves the quality of the obtained spectra for the range of emergency radiation doses, even taking into account different types of spectrometers on which measurements are carried out.

Keywords: *EPR-dosimetry, tooth enamel, EPR microwave power, emergency human dosimetry, EPR spectra.*

INTRODUCTION

Radiation accidents, both large-scale and local, can occur as a result of a combination of various factors, including careless in handling radioactive materials or difficulties associated with the operation of radioactive sources at industrial facilities of the nuclear industry [1–3]. In the event of a radiation accident, information about the radiation dose of an irradiated patient is necessary for medical treatment and triage of an irradiated patient [4]. EPR dosimetry using biological samples of an irradiated patient (teeth, nails and hair) and his personal items has been widely studied for several decades for dose assessment. Tooth enamel is a well-known sample for assessing the effects of radiation on a patient in a radiological accident.

In the past, the method of EPR dosimetry of tooth enamel was actively investigated and used for the retrospective assessment of emergency doses after large-scale incidents such as the accidents at the Chernobyl nuclear power plant, Fukushima and for assessment of consequences of SNTS activity (Kazakhstan).

Currently, it is known that the method of tooth enamel EPR dosimetry is relevant for emergency radiation dosimetry, due to the high sensitivity of samples to high doses. Recent studies include such issues as the development of techniques for preparing samples without removing a whole tooth, by using only a tooth enamel biopsy [5, 6]. Also the approaches for different spectrometers to obtain a "purified" radiation-induced signal (RIS) are being studied [7, 8]. Various approaches to optimizing the procedure for calculating and interpreting EPR spectra are also being investigated, such as estimating the minimum detection limit of RIS depending on the mass of the sample [9].

Despite the fact that EPR dosimetry of tooth enamel is currently being more actively developed in world science, especially in the field of emergency radiation doses, over the past 10 years, the experience of using this method in Kazakhstan has been limited by monitoring and calculating chronic retrospective doses in residents of the vicinity of the SNTS territory [10–12]. Then, there is one known study on the assessment of chronic doses received by employees of Kazakhstan's uranium processing plant [13]. However, in all these works, the EPR technique is applied according to the standard protocol without significant modifications of the method.

It should be noted that the application of the EPR dosimetry method for human tooth enamel, currently in Kazakhstan, has no research experience in modifying a standardized technique or improving the quality of the procedure for calculating EPR spectra, with the exception of one work [14].

In 2018, a study was conducted to optimize the parameters of spectrum measurement for tooth enamel EPR spectroscopy in order to improve the accuracy of the method based on a domestic laboratory [14]. As a result, the authors developed an approach to optimize the parameters of EPR spectra registration by determining the most effective range of microwave power values and the accumulation time during registration of spectra.

The authors determined the minimum uncertainty limit for EPR spectra with different microwave power – 2 MW, and also analyzed the effectiveness of increasing the accumulation time.

However, in this study, the authors used a range of low radiation doses (up to 0.5 Gy). The optimization of the specifically described parameters in this work was due to the dependence of the accuracy of experimental dose calculation with special recording parameters.

Nevertheless, it is known that the quality and reliability of dose calculation in a range of high doses (from 0.5 Gy) also depends on particular EPR spectra registration parameters [15, 16]. But for each individual study, it is necessary to determine individual values, considering the range of experimental doses and particular spectrometer features.

The purpose of our study was to develop a modification of recording parameters for human tooth enamel EPR spectra samples, such as microwave power (microwave) under conditions of a local radiation incident based on the biodosimetry laboratory of Kazakhstan.

MATERIALS AND METHODS

Sampling was carried out from 3 adult donors of the same age range (35–40 years old), which are residents of Ust-Kamenogorsk. The selected age range was chosen due to the influence of the donor's age on the quality of the measured EPR signal, since it depends on the number of the tooth enamel paramagnetic centers. This number increases throughout an individual's life. Up to 30–35 years, human tooth enamel may accumulate an insufficient number of centers, while after 45 years there is an excess of them, which can significantly contribute to parasitic noises and background RIS signal. Based on this, a specific age range was selected in this study.

A total sum of 10 teeth removed for medical reasons at a dental clinic were included in the analysis. The donors did not live in radioactively contaminated areas and were not exposed to acute emergency or chronic radiation exposure. According to information obtained through a donor questionnaire, dental samples have not been subjected to X-ray irradiation in the jaw area in the last 6 months.

Due to the special complexity and complexity of the sample preparation procedure, it was decided to conduct a study on combined samples. In addition, this study does not search the effect of donor age on the quality of the EPR signal due to the homogeneity of the group.

The selected samples were divided into buccal and lingual parts. The position of each sample was also taken into account in order to avoid ultraviolet sunlight exposure, since they may provoke the appearance of additional parasitic signals in RIS.

The preparation of combined samples of powdered tooth enamel was carried out according to the standard method of chemical treatment of whole teeth samples. After chemical treatment, the enamel was crushed into particles of 0.5–1.5 mm in size. This granule size was chosen because it is the most optimal in terms of minimizing the effects of sample anisotropy and surface noise, which leads to distortion of the EPR signal. Mixed samples were divided into portions equated to 100 mg, and then irradiated. According to the results of the sample preparation, 3 samples of 73.6, 77 and 75.4 mg were obtained, respectively.

The irradiation was carried out at a gamma-ray installation with a source of Cs^{137}. The samples were also irradiated at 3 dose points: 1.3 and 5 Gy, and measurements at the 0 Gy point were also included. This range of dose points was determined due to the special effectiveness of the investigated method in measuring high emergency doses. Since this is a pilot study on the modification of measurement parameters, it was decided to choose a small range of dose points in order to avoid strong uncertainties when measuring EPR spectra. In further studies, it is planned to use a wider range of experimental radiation doses. Dose control was carried out with preliminary measurement of the enamel spectra before irradiation, then equated to the sample spectrum of manganese.

All measurements were performed in the X frequency range on a Bruker ESP 300 E EPR spectrometer at a stabilized room temperature of 21 °C. The spectrometer is equipped with a spherical TE ES-UCX2 resonator with a high Q factor. Table 1 shows the parameters for measuring the spectra.

Table 1. Parameters for measuring EPR spectra on tooth enamel samples

Name, unit	Value
The amplitude of the signal intensity, MT	3622
Field width, MT	3480
Scan width, MT	93.1
Modulation frequency, kHz	100
Receiver of the time constant, ms	20.48
Scan time, ms	40.96
Frame scan, Mt	10
Microwave power, MW	9.70
Number of scans	50
Spectrum set time, min	5–20

The signal of the control sample containing Mn^{2+} installed in the cavity was recorded together with each sample spectra. The signal in the tube spectrum was received with a minimum value, and no subtraction was carried out.

RESULTS AND DISCUSSION

For computer processing of EPR spectra, an automated program was used as part of the spectrometer software. This procedure is capable of scanning the experimental spectrum of irradiated enamel using the nonlinear least squares method using a model spectrum describing an analytical form of RIS and BGS (background signal). The spectrum processing procedure was applied in the BGS description mode with two components consisting of a combination of derivatives of Gaussian functions.

The window was set with a left boundary of −1.0 MT and a right boundary of +2.0 MT relative to the BGS maximum. During the fitting process, the following parameters were changed: RIS amplitude, amplitudes of narrow and wide BGS components, width of the narrow

BGS component, vertical and horizontal shifts of the spectra. The baseline correction was performed by fitting a first-order polynomial to two regions of the spectrum between the tooth enamel signal and two Mn^{2+} signals.

At the same time, RIS amplitude was determined for a series of spectra by irradiated samples at different doses and measured with the same registration parameters.

Then, according to the dependence of the RIS amplitude on the nominal dose, the parameters of the calibration dependence were determined by linear regression and, using these parameters, the dose in the enamel was determined based on RIS amplitude.

To optimize the parameter for recording spectra, EPR signals from the spectra of 3 samples weighing up to 100 mg were recorded at 10, 20, 40 scans (which corresponds to an accumulation time of 5, 10, 20 minutes, respectively) and at microwave power of 1, 5 and 15 MW.

Figure 1 shows that RIS amplitude, depending on the increase in the experimental dose, has a linear structure for all 3 samples at a microwave power value of 5 MW. This is due to an increase in both low-frequency and high-frequency noise components in the spectra at other parameter values.

There is a comparative uniformity of the signal increase with an increase in microwave power at dose points 1 and 3 Gy (Fig. 1a; 1b) however, at the dose point of 5 Gy, a deviation in the curve was recorded at a power value of 5 MW. The presence of such fluctuations is probably due to the influence of the heterogeneity of the RIS in the experimental doses values, since at this dose point EPR signals for all samples show a sufficient range of values. We suppose that it can be caused by increased noise component at a high radiation dose, as well as specific spectrometer characteristics.

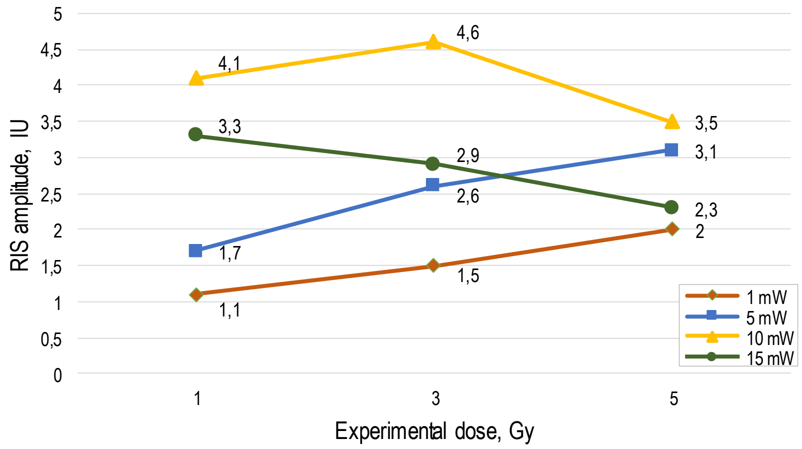

Figure 1. Sample 1 – Dependence of the RIS amplitude on range of the microwave power with 3 dose points

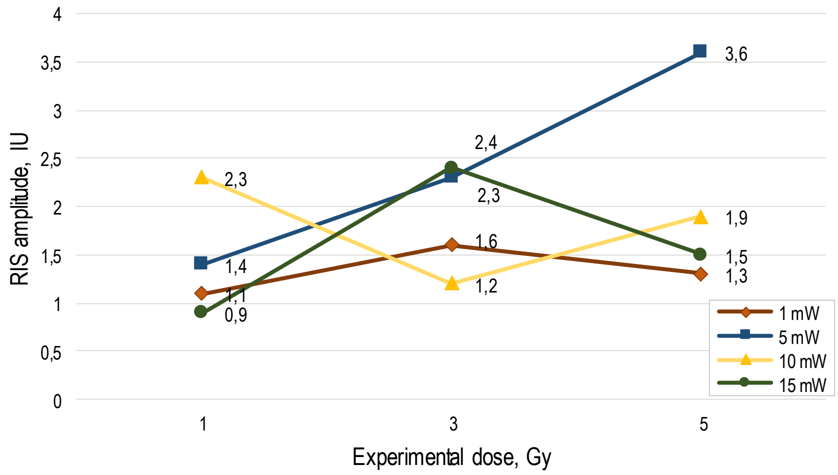

Figure 2. Sample 2 – Dependence of the RIS amplitude on range of the microwave power with 3 dose points

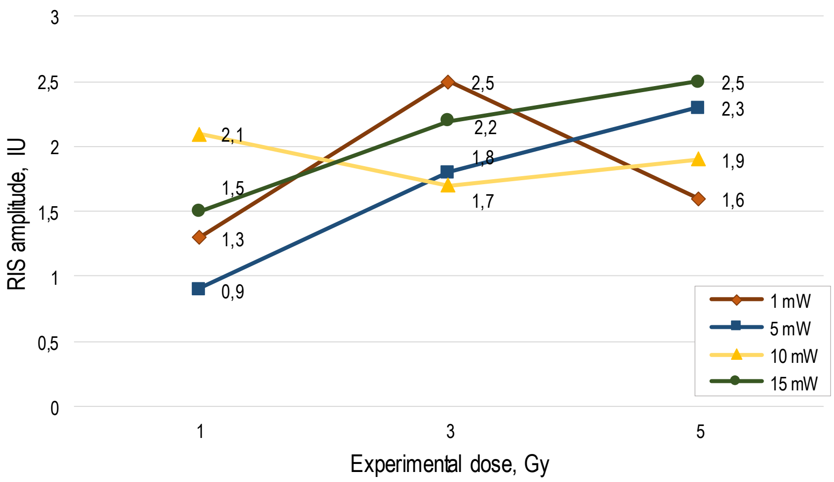

Figure 3. Sample 3 – Dependence of the RIS amplitude on range of the microwave power with 3 dose points

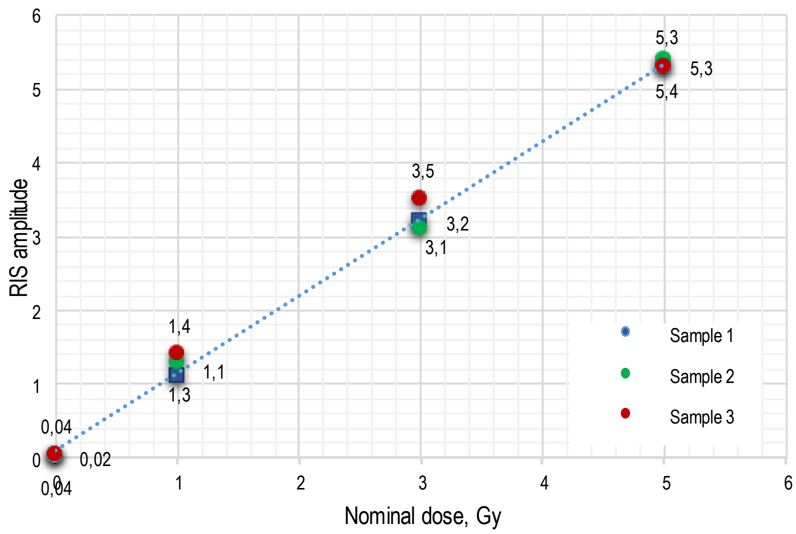

Figure 4. The obtained experimental values of all samples at 3 dose points

In comparison with the previous work [15], there is a difference in a certain effective value of microwave power for the most reliable calculation of the absorbed experimental dose. It has been established that the most effective value of microwave power for recording EPR spectra in the range of emergency doses is 5 MW.

The slope curve of the experimental dose values from the nominal ones was also calculated. The experimental absorbed dose accumulated in the samples was calculated using a standard formula.

As a parameter characterizing the accuracy of dose determination, for each series of spectra measured with the same registration parameters, the standard deviation of the dose between certain and nominal values (SDD) was calculated.

As it can be seen from Table 2, the values of experimental doses vary slightly from the nominal values. However,standard deviation for each sample correspond to the literature data and generally accepted meanings for tooth enamel samples. Next Figure 4 demonstrates the spread of experimental dose values relative to the nominal curve.

Table 2. Calculated values of the received doses

Nominal dose, Gy	Sample, №	Experimental dose, Gy	SDD, %
0	1	0.02	10
	2	0.04	
	3	0.04	
1	1	1.1	13
	2	1.3	
	3	1.4	
3	1	3.2	16,6
	2	3.1	
	3	3.5	
5	1	5.3	19
	2	5.4	
	3	5.3	

Figure 4 shows the increasing of the RIS peaks. With regard to the curve of the energy dependence of the intensity values of tooth enamel RIS in relation to experimental dose, we can conclude, that this parameter has a fairly linear shape (Figure 4). However, there are discrepancies in the dose range from 0.4 to 5.8 Gy (the values of the actual absorbed doses). These differences can be explained by the presence of background noise signals that somehow occur with each repeated measurement, as well as possible technical features of the spectrometer used. This is also due to the fact that even on the same tooth, the degree of formation of stable radicals depends on the chemical structure of each enamel sample.

A study of the EPR spectroscopy approach for human tooth enamel was conducted, taking into account the optimization of the parameter for recording EPR spectra (microwave power) at 3 dose points under conditions of a local radiation incident.

It was found that at a microwave power value of 5 MW, a linear increase in the EPR signal is observed, similar to the amplitude of the RIS with increasing dose.

It is shown that the obtained results correlate with the literature data on the use of this modification in the field of low doses of radiation. The values of the standard deviation were expected and do not conflict with the standardized methodology.

It was found that the increased differences in the intensity values of RIS at the dose point of 5 Gy, depending on the increase in microwave power, can be explained by the influence of a noise signal, as well as the uncertainty of the signal at the highest radiation dose.

CONCLUSION

In this study, we applied an optimization technique for the parameter of recording EPR spectra on human tooth enamel, in the range of emergency radiation doses (from 1 Gy to 5 Gy), under conditions of a local radiation incident. The results showed that optimization of the parameter for recording EPR spectra, in particular microwave power, improves the quality of the obtained spectra for a range of emergency radiation doses, even taking into account different types of spectrometers on which measurements are carried out.

The further direction for research should be the improvement of approaches for dose calculation EPR methods in the range from 1 to 3 Gy, as well as the study of other methods for optimizing processing parameters and calculating EPR spectra for emergency doses.

The application and study of individual EPR spectrometry and physical dosimetry techniques for the identification and evaluation of the absorbed dose specifically in the range from 1 Gy represents an extensive promising area for applied research, due to high individual sensitivity and reliable results, which are the primary factors of high-quality individual dosimetry.

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АДАМНЫҢ АПАТТЫҚ ДОЗИМЕТРИЯСЫ ҮШІН ЭПР-СПЕКТРЛЕРДІ ӨЛШЕУ ПАРАМЕТРЛЕРІН ОҢТАЙЛАНДЫРУ ӘДІСІН ҚОЛДАНУ

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Әлемдік әдеби деректерден 0,5 Гр және одан жоғары дозалар пайдаланылатын апаттық дозиметрия мақсатында ЭПР әдісін пайдаланған кезде эксперименттік дозаны есептеудің сапасы мен дұрыстығы спектрлерді тіркеу параметрлеріне (жинақтау уақыты, микротолқынды қалындық) байланысты екені белгілі.
Бұл мақалада жергілікті радиациялық оқиға жағдайында адамның тіс эмальында ЭПР спектрлерін тіркеу параметрін оңтайландыру әдісін әзірлеу және қолдану нәтижелері сипатталған. Зерттеу 1 Гр-ден 5 Гр-ге дейін доза диапазонында сәулеленген адамның тіс эмальының үлгілерінде жүргізілді. Нәтижелер 5 МВт микротолқынды қалындығында апаттық сәулелену дозалары диапазоны үшін адамның тіс эмальының ЭПР спектрлерінің сапасын жақсартудың ең тиімді параметрі екенін көрсетті. ЭПР спектрлерін тіркеу параметрін оңтайландыру, атап айтқанда микротолқынды қалындығы, өлшеу жүргізілетін спектрометрлердің әртүрлі түрлерін ескере отырып, апаттық сәулелену дозалары диапазоны үшін алынған спектрлердің сапасын арттырады.

Түйін сөздер: *ЭПР-дозиметрия, тіс эмальы, микротолқынды қуат, физикалық дозиметрия, ЭПР-спектр.*

ПРИМЕНЕНИЕ МЕТОДА ОПТИМИЗАЦИИ ПАРАМЕТРОВ ИЗМЕРЕНИЯ ЭПР-СПЕКТРОВ ДЛЯ АВАРИЙНОЙ ДОЗИМЕТРИИ ЧЕЛОВЕКА

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Из мировых литературных данных известно, что при использовании метода ЭПР в целях аварийной дозиметрии, где используются дозы от 0,5 Гр и выше качество и достоверность расчета экспериментальной дозы зависит от параметров регистрации спектров (время накопления, микроволновая мощность).
В данной статье описываются результаты разработки и применения метода оптимизации параметра регистрации спектров ЭПР на зубной эмали человека, такого как микроволновая мощность, но в диапазоне аварийных доз облучения (от 1 Гр до 5 Гр), в условиях локального радиационного инцидента на базе биодозиметрической лаборатории. Результаты показали, что полученное экспериментальное значение мощности СВЧ 5 МВт является наиболее эффективным параметром для повышения качества ЭПР-спектров зубной эмали человека для диапазона аварийных доз облучения, даже с учетом разных типов спектрометров, на которых проводятся измерения. Оптимизация параметра регистрации спектров ЭПР, в частности мощность СВЧ, повышает качество полученных спектров для диапазона аварийных доз облучения, даже с учетом разных типов спектрометров, на которых проводятся измерения.

Ключевые слова: *ЭПР-дозиметрия, зубная эмаль, микроволновая мощность, физическая дозиметрия, ЭПР-спектр.*