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A TRANSFERABILITY STUDY OF THE EPR-TOOTH-DOSIMETRY TECHNIQUE

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The transferability of a measurement protocol from one laboratory to another is an important feature of any mature, standardised protocol. The electron paramagnetic resonance (EPR)-tooth dosimetry technique that was developed in Scientific Center for Radiation Medicine, AMS, Ukraine (SCRM) for routine dosimetry of Chernobyl liquidators has demonstrated consistent results in several inter-laboratory measurement comparisons. Transferability to the EPR dosimetry laboratory at the National Institute of Standards and Technology (NIST) was examined. Several approaches were used to test the technique, including dose reconstruction of SCRM–NIST inter-comparison samples. The study has demonstrated full transferability of the technique and the possibility to reproduce results in a different laboratory environment.

INTRODUCTION

Despite three international interlaboratory comparisons[1–3] and an International Atomic Energy Agency (IAEA)-coordinated project[4] with participation of many electron paramagnetic resonance (EPR)-dosimetry laboratories, a standardised EPR-dosimetry technique for tooth enamel has not yet been resolved. All EPR-tooth-dosimetry techniques currently in use have certain aspects of them that are different. Since the uncertainties cited by the originators of the various techniques are always combined, it is not possible to assess and compare the quality of the different approaches so that a unified protocol can be developed.

An alternative approach is to select one of the dosimetric techniques that performed well in the inter-comparisons, as the basis for a standardised technique from which improvements can be made. The first step in this approach is to examine the reproducibility, without loss of quality, to another dosimetric laboratory. The purpose of this study is to report the transfer of the EPR-dosimetry technique developed and routinely used at Scientific Center for Radiation Medicine, AMS, Ukraine (SCRM) to the EPR-dosimetry laboratory of National Institute of Standards and Technology (NIST).

MATERIALS AND METHODS

General characteristics of the technique to be reproduced at NIST

SCRM technique consists of the following steps.

Step 1: preparation of samples for EPR-spectra registration

Sections are cut (~2–3 mm) from lingual and buccal sides of a tooth (lateral teeth with size 8–10 mm) or a whole tooth is cut into two halves (lateral teeth with size 5–7 mm). Front teeth (incisors and canines) are not used for dosimetry because of dosimetric interferences from ultraviolet solar exposure. The dose reconstruction procedure is conducted separately for the lingual and buccal sides.

Tooth enamel is purified from dentine and surface contamination using mechanical or chemical methods (or combination of two methods in rare cases). The mechanical method is used when the number of samples to be processed is small and it is desirable to reconstruct doses as rapidly as possible. The mechanical method is also applied if it is impossible to use the chemical method. The chemical method is preferable in case of a large number of samples or when samples after mechanical separation demonstrate strong impurity signals in EPR spectra. The quality of the two methods is approximately the same within the limits of the SCRM's technique: according to special tests, the value of the dosimetric signal does not depend on the sample preparation method.

After separation, the enamel sample is washed in ethanol, dried at 90°C for 2 h and crushed to 0.5–1 mm particles using dental pliers. A 90–100 mg aliquot is taken from the sample for EPR-spectra registration.

Step 2: measurement of EPR spectra

The aliquot of the sample is put into a 5 mm quartz tube and inserted into the EPR-microwave cavity.

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A Mn^{2+} MgO reference sample is also inserted into the cavity through the bottom hole. The following parameters are used for spectra registration: 10 mW microwave power, 0.4 mT modulation amplitude, 100 kHz modulation frequency, 10 mT field sweep, 20 ms analog-to-digital conversion time, 20 ms filter time, 120 scans and 1024 channel resolution.

The spectrum of each sample is recorded using a programmable goniometer at 10 different angles (with respect to constant magnetic field direction) to reduce the anisotropy effect. The spectra of the empty sample tube are recorded in the same manner; these spectra are used for correction of sample spectra. The measurement cycle for one sample consists of following sequence:

- measurement of the empty sample tube spectrum: the summation of 10 spectra with 6 scans each, rotation of tube with sample inside with tilt of 18° between two serial spectra recordings;
- measurement of sample spectrum: the summation of 10 spectra with 12 scans each, the same rotation as above; and
- repeated measurements of the empty sample tube spectrum.

The spectrum measurement cycle for laboratory-irradiated samples (see Step 4 below) is simpler: only three scans are accumulated at every angle and only one set of empty sample tube spectra (3 scans, 10 angles) is recorded during one working session. This simplification is possible because daily variations of empty sample tube spectra are insignificant compared with the very strong dosimetric signal in laboratory-irradiated samples.

The spectra are processed by means of a deconvolution algorithm to estimate the dosimetric signal in the sample.

Step 3: deconvolution of spectra

The contribution of the dosimetric signal in the EPR spectrum is determined by a deconvolution procedure, which uses some experimental spectra as standards of the spectral components. A detailed description of the SCRM's deconvolution algorithm is given in Sholom and Chumak[5]. It is necessary to note that the most critical element of this procedure is the selection of the standard spectra. It was found in Ref. (5) that it is sufficient to use three standard spectra (after subtraction of empty sample tube spectrum) to describe the EPR-spectra of tooth enamel samples of Ukrainian origin. In this work, both Ukrainian and US teeth were cross-analysed using the two spectrometers; therefore new standard spectra were created separately for teeth of different origins. For this purpose, five presumably unexposed Ukrainian teeth and the same number of US teeth were used as described in Ref. (5).

Step 4: calibration of EPR signals in units of absorbed dose

The SCRM technique calibrates radiation sensitivity of each tooth sample by applying a calibrated dose to a portion of the sample in the range 5–10 Gy. The strong EPR signal resulting from this dose enables the tooth-dose sensitivity to be determined with high precision (within 1%). After irradiation, samples are annealed at 90°C for 2 h to remove short-lived EP resonances[6].

The absorbed dose of the sample is determined as the product of two variables: the value of the dosimetric signal in the initial sample and the sample's radiation sensitivity.

Step 5: estimation of dose component due to accidental exposure

The absorbed dose in enamel is a sum of three components[7]: the dose owing to natural background radiation, the X-ray diagnostic dose and the 'accident' dose component (the ultraviolet solar exposure component is considered to be negligible for lateral teeth). The most uncertain component is because of the possible X-ray diagnostic exposures. This component may be accounted for using separate measurements of lingual and buccal tooth sections, as described in Ref. (8). Though normally contribution of the natural background dose is accounted by multiplication of annual dose rate by the age of tooth (this parameter depends on the age of the subject and type of tooth)[7], in this study teeth from young subjects were used and possible dose (not more than several mGy) was neglected.

So, the accidental dose in enamel is estimated as indicated above.

Comparison of available facilities[a]

The main components of the EPR-dosimetry facilities that are available in the two laboratories are listed in Table 1.

The NIST laboratory is equipped with a higher-quality EPR spectrometer and a cavity (Bruker ER4122 SHQ) with highest Q-value available. This permits a much shorter period of time to achieve the same 'dosimetric signal-to-noise' ratio as for ECS106 spectrometer with TMH 4108 cavity. In most other respects, the differences between the facilities of the two laboratories are insignificant.

[a]The mention of commercial products throughout this paper does not imply recommendation or endorsement by the NIST nor does it imply that products identified are necessarily the best available for this purpose.

Samples used in the study

Three sets of samples were used. The first set consisted of the 10 already mentioned unexposed teeth used to produce both native (samples without any additional irradiation) and dosimetric (the same samples after 10 Gy laboratory irradiation) standard signals. The second set included several samples exposed at NIST in the dose range 50–1000 mGy and used in the reproducibility study and for some other tests. The third set consisted of 10 teeth exposed at IAEA in 2002 in the framework of bilateral SCRM–NIST inter-comparison in the dose range 0–300 mGy. After irradiation all teeth were cut in two halves in such a way that every half comprises both lingual and buccal parts: one set of halves was analyzed at SCRM, and the other at NIST.

Table 1. Facilities of SCRM and NIST laboratories used for EPR dosimetry with teeth.

Component of facilities	Availability in SCRM	Availability in NIST
EPR spectrometer	Bruker ECS106	Bruker Elexsys
Microwave cavity	Bruker TMH 4108	Bruker SHQ 4122
Programmable goniometer	Bruker ER 218 PG1	Bruker ER 218 PG1
Low speed diamond saw	Buehler	Buehler
Ultrasonic bath	Bransonic	Bransonic
Dental drill machine	+	+
Oven for sample drying	+	+
Dental pliers	+	+
500–850 μm sieves	+	+
PC (Pentium II class)	+	+
^{137}Cs gamma source (%) (95% Confidence)	3[a]	1.5

[a]The SCRM source dose rate is traceable to NIST by alanine transfer dosimetry[9]

RESULTS AND DISCUSSION

Optimal parameters of spectra registration for NIST spectrometer

Power dependences were obtained for TMH ER4108 (SCRM spectrometer) and SHQ ER4122 (NIST spectrometer) cavities. Using these dependences the optimal value was determined to be 10 mW for TMH cavity and 2 mW for the SHQ cavity. From the 'signal-to-noise' ratio study it was found that the number of spectral scans for the Elexsys spectrometer may be reduced by two without affecting the optimal 'signal-to-noise' ratio. Other parameters of spectra registration on Elexsys spectrometer were the same as for the SCRM ECS106 spectrometer.

Standard spectra obtained for NIST technique

Standard spectra determine the central element in the SCRM technique of spectra deconvolution. It was found that it is sufficient to use only two standard spectra (after subtraction of empty sample tube spectrum) to fit successfully the enamel sample spectra. Although the standard spectra for the dosimetric signal standards were approximately the same, the native signal standards slightly differed for the US and Ukrainian teeth (Figure 1). The parameters for the native signal standards were: *g*-factor values

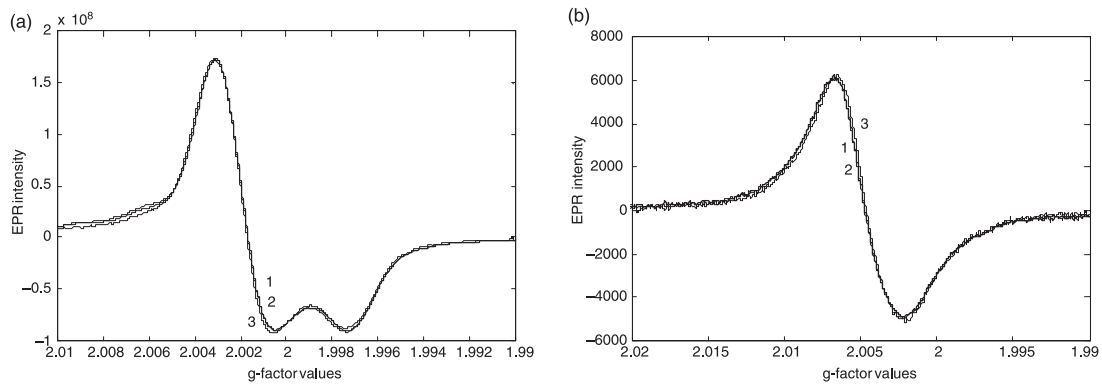

Figure 1. Standard reference spectra used at SCRM and at NIST. Plot a shows standard spectra of the dosimetric signal, plot b standard spectra of the native signal. Numbers 1–3 correspond to the following spectra: 1, spectra used at SCRM; 2, spectra used at NIST for Ukrainian teeth; 3, spectrum used at NIST for US teeth.

2.0045 and 2.0046 (±0.0001), peak-to-peak width 0.83 and 0.85 (±0.01) mT for US and Ukrainian teeth, respectively. These values are close to those observed by other authors [compare with 2.0046 and 0.833 mT in Ref. (10) or 2.0045 and 0.78 mT in Ref. (11)].

An additional signal in the standard spectrum that was observed in Ref. (5) was not observed in this work because this signal is probably related to the chemical treatment of samples, which was not applicable in this study.

Testing of the reproduced technique: measurements of reference samples

Measurements were done in two stages.

Stage 1: study of reproducibility of dosimetric signal measurement in samples exposed to 50, 100, 200 and 1000 mGy. The results obtained for both spectrometers are presented in Table 2. Reproducibility was determined from measurements in triplicate for the corresponding samples; careful shaking of the tube with sample inside was done between measurements. The absorbed doses in Table 2 are obtained by signal normalisation to a 1 Gy sample. The reproducibility of EPR signals was better for more sensitive Elexsys spectrometer, especially for low dose region.

Stage 2: dose reconstruction for samples irradiated in the 50–200 mGy range. The results of this test are shown in Table 3. It should be noted that for this particular test the dose of an unexposed aliquot of a tooth was used as a 'background' dose for its corresponding irradiated sample. This background dose includes both the natural background and the X-ray dose components. This approach resulted in very small (within 10 mGy) deviations of reconstructed doses from the corresponding nominal values for all samples.

Table 2. Results of reproducibility experiment (series of 3 measurements) for dosimetric signal.

Nominal dose	50 mGy	100 mGy	250 mGy	1000 mGy
NIST results				
Mean value (mGy)	51	93	261	1000
SD (mGy)	11	2	17	9
SD (%)	21	2	6	0.9
SCRM results				
Mean value (mGy)	56	118	267	1000
SD (mGy)	14	13	22	15
SD (%)	25	11	8	1.5

Dose reconstruction for inter-comparison samples

Results of dose reconstruction of 10 inter-comparison teeth are presented in Table 4. All samples can be assigned (conditionally) to three groups. The first group includes all samples of Ukrainian origin as well as two samples (C and F) from USA. For this group both techniques demonstrated approximately the same deviation of reconstructed doses from nominal values. The second group consists of US samples J and H. For this group, NIST results are better, because a native signal standard for US teeth was used. This assumption is demonstrated in Figure 2 in which an example of spectral fitting using different standards (obtained on US teeth in case of plot a and on Ukrainian teeth in case of plot b) is shown for sample H. The quality of the fit is better when the US tooth standard is used. It should be noted that dose detected in sample H was several tens of milligray when Ukrainian standard was used and ~0 in case of US standard. The third group consists of one sample B for which NIST and SCRM reconstructed doses were practically the same (394 and 377 mGy), but significantly higher than the corresponding assigned value (168 mGy). In our opinion, of the possible causes (impurity signals, sample preparation and dental X-ray exposure) the most probable reason for this deviation is the presence of an X-ray dose (possibly from a combination of multiple local and panoramic X-ray examinations) in sample B. The sample B result is considered an outlier and excluded from further analyses.

The measurement correlation (Figure 3) has been plotted using the nine samples from the NIST study and five Ukrainian samples from the

Table 3. Results of dose reconstruction for reference samples at NIST.

Absorbed dose (mGy)	EPR dose (A) mGy)	'Background' dose (B) (mGy)	Reconstructed nominal dose (A – B) (mGy)	Deviation of reconstructed from nominal dose (mGy)	Deviation in %
200	211	14	197	3	2
100	145	40	105	5	5
50	99	40	59	9	18

Table 4. Results of dose reconstruction for SCRM-NIST inter-comparison study.

Sample no. (country of origin)	Nominal (IAEA) dose (mGy)	NIST dose (mGy)	Deviation of NIST from IAEA doses (mGy)	SCRM dose (mGy)	Deviation of SCRM from IAEA doses (mGy)
A (Ukraine)	128	115	−13	146	18
B (USA)	168	394	226	377	209
C (USA)	236	203	−33	259	23
D (Ukraine)	195	156	−39	176	−19
E (Ukraine)	269	276	7	279	10
F (USA)	275.5	251	−24.5	280	4.5
G (Ukraine)	0	0	0	22	22
H (USA)	0	3	3	77	77
I (Ukraine)	164	144	−20	160	−4
J (USA)	118	117	−1	202	84

Grey background cells indicate outlier or improper values (see details in text) which were excluded from correlation analysis

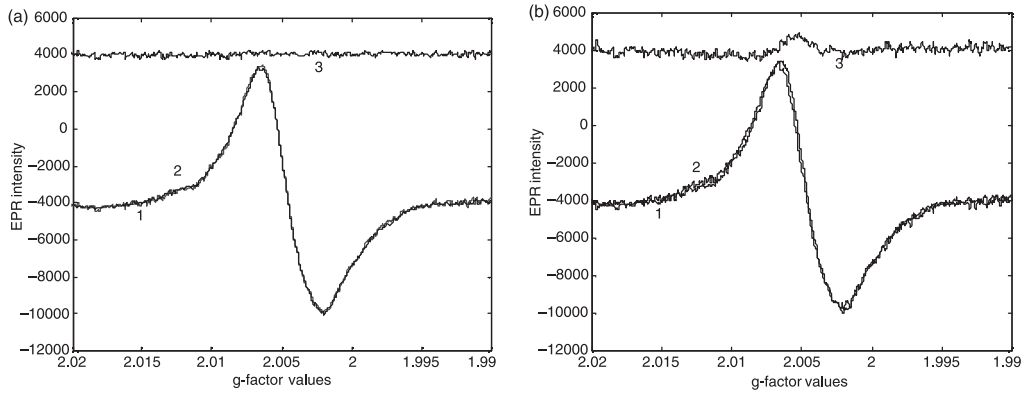

Figure 2. Example of spectra fitting (sample H, see Table 3) using different native signal standards: (a) obtained on US teeth and (b) on Ukrainian teeth. Lines 1–3 are: 1, initial spectra; 2, fitting spectra; and 3, difference between 1 and 2.

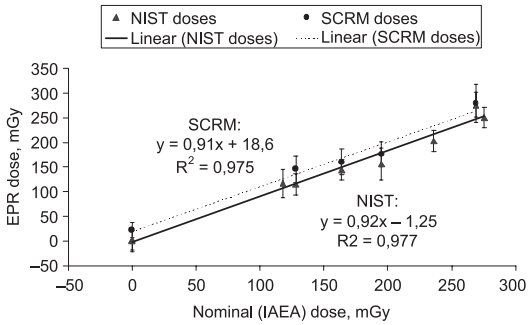

Figure 3. Correlation of NIST and SCRM reconstructed doses with nominal values.

SCRM study (US teeth were excluded from SCRM study because no native signal standard for US teeth was available). The results from both laboratories are comparable: the NIST results had an SD of 21 mGy and a correlation coefficient 0.977; the SCRM had an SD of 16 mGy and a 0.975 correlation coefficient.

CONCLUSION

The EPR-dosimetry technique that was developed and implemented at SCRM was successfully transferred to NIST. The spectrometer parameters for the technique corresponded to parameters of the original one, but were slightly better because of higher sensitivity of NIST's spectrometer. It was found (as a result of dose reconstruction for inter-comparison teeth) that the differences between reconstructed dose and actual absorbed dose do not exceed 39 mGy with an SD of 21 mGy.

These results offer an optimistic outlook for the development of a standardised EPR-tooth-dosimetry technique destined for a broader circle of users.

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