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EPR and OSL emergency dosimetry with teeth: A direct comparison of two techniques

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HIGHLIGHTS

- OSL and EPR dose reconstruction techniques were compared on the same teeth.
- Values of minimum measurable doses for the OSL technique were in the range 0.9–1.5 Gy.
- Fading of OSL signals correlated with fading of EPR signals attributed to CO_3^- radicals.

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ABSTRACT

The OSL dosimetry technique with teeth has been validated against the EPR dosimetry technique through a measurement comparison performed on the same teeth. The OSL reconstructed doses were found to be in agreement with corresponding EPR doses. Minimum measurable doses for the OSL technique were estimated to be in the range 0.9–1.5 Gy for measurements made within 24 h post-exposure if OSL signals are collected from 6 teeth at the same time. These values satisfy the requirements for emergency triage dosimetry. The fading of tooth OSL signals correlated with fading of radiation-induced EPR signals observed at $g = 2.0115$ that are attributed to CO_3^- radicals. OSL sensitivity can be enhanced if more teeth from the same individual will be used for the signal accumulation.

1. Introduction

Tooth enamel is well established as a reliable Electron Paramagnetic Resonance (EPR) dosimeter. It was used for the dose reconstruction of A-bomb victims in Japan (Ikeya et al., 1984); Chernobyl Nuclear Power Plant liquidators in Ukraine and Russia (Chumak et al., 1999; Skvortsov et al., 2000); Mayak Nuclear Facility workers in Russia (Romanyukha et al., 1994); Techa river population in Russia (Romanyukha et al., 2001); and the Semipalatinsk Nuclear Test Site region population in Kazakhstan (Sholom et al., 2007). The main advantage of the tooth EPR dosimetry technique is the high stability of the enamel dosimetric EPR signal, which is mainly attributed to CO_2^- radicals (Vanhaelewyn et al., 2002). The half-life of these radicals at ambient temperature is believed to be hundred thousands of years (Schwarcz, 1985). This stability combined with a radicals' high yield and the sensitivity of the modern EPR spectrometers, enables dose reconstruction at levels as low as 50–100 mGy many days after an accidental exposure (Wieser et al., 2006).

The drawback of traditional tooth EPR dosimetry techniques is their "in vitro" applicability, i.e. they work only with extracted teeth. Adaptations of the technique for "in vivo" (in a L-band of microwave frequency, see e.g. Swartz et al., 2012) or "quasi-in-vivo" (in a Q-band of microwave frequency, see e.g. Romanyukha et al., 2007) dose reconstruction have been explored, but remain in the development stages and pending further verifications before practical use.

Another possibility for in-vivo dose reconstruction with teeth is the Optically Stimulated Luminescence (OSL) technique (DeWitt et al., 2010). Significant progress has been achieved with this technique during last two decades: values of minimum measurable dose (MMD) have been reduced from about 120 Gy to doses below 0.64 Gy for samples measured immediately after exposure (Godfrey-Smith and Pass, 1997; Yukihara et al., 2007; Godfrey-Smith, 2008; DeWitt et al., 2010; Sholom et al., 2011), which is an acceptable level for triage applications. But due to the strong fading

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of OSL signals in teeth, a question regarding the reliability of doses reconstructed with the OSL technique is still open. The tooth doses reconstructed with OSL measurements have never been checked against an independent dosimetry technique. Also, there has been no experimental validation for MMD values and their corresponding uncertainties for samples measured a day or more after exposure.

In the current study, the OSL dosimetry technique with teeth has been validated against an established EPR dosimetry technique using the same teeth. MMD values have been determined at different times after the sample's exposure. Some assumptions about nature of OSL signals from teeth have been suggested.

2. Materials and methods

The dose reconstruction was conducted on a set of 30 teeth (molars) extracted for medical reasons at local dental clinics without any individual information. The teeth were cut in halves with a Buehler Isomet low speed diamond saw, with the intent that one set of tooth halves would be tested by the EPR technique and another set of halves measured by the OSL technique. The 30 teeth were irradiated to a 5 Gy dose using a NIST-calibrated ^{60}Co gamma-ray source; this dose was used to simulate an emergency exposure.

EPR dose reconstruction was carried out using a Bruker Elexsys EPR spectrometer equipped with a Bruker 4119 cavity and operating in the X-band. The spectral recording parameters were: microwave power 12.7 mW; field sweep 10 mT; modulation amplitude 0.4 mT; conversion time 20 ms; time constant 20 ms; spectral resolution 1024 data points. Thirty aliquots of tooth enamel in the weight range between 27 and 102 mg were prepared from corresponding halves of teeth using a low speed dental drill. These aliquots were used for EPR measurements without any chemical treatments. Each of the thirty samples was measured at 10 different angles with respect to the direction of the permanent magnetic field to reduce the possible influence of EPR signal anisotropy. For the calibration of EPR signals, samples were irradiated to a 10 Gy dose and measured again. All spectral fitting and analysis was done with MatLab software according to the methods of Sholom and Chumak (2003).

No sample preparation was applied to the tooth halves used for OSL measurements. A group of six halves were combined into a multi-tooth sample; and the total number of these samples was five. This was done as described in Sholom et al., 2011 to increase the signal-to-noise ratio. OSL measurements were conducted using a custom OSL reader; samples were stored in the dark during all time between irradiation and OSL readout to avoid the bleaching of OSL signals by environmental light. Samples were stimulated with Blue (470 nm) light-emitting diodes. Emission from the samples was detected by a photomultiplier tube after passing Hoya U-340 UV filters (290–390 nm). The duration of the stimulation was 30 s with a spectral resolution of 100 ms per data point. OSL dose reconstruction was conducted 1 day after irradiation to simulate a possible triage application. For the calibration of OSL signals, samples were irradiated to a 16 Gy single dose using an X-ray source operated at 120 kVp and 0.64 mm Al filtration and calibrated against the NIST ^{60}Co source by using teeth as transfer dosimeters; all doses reported in the current study are expressed in ^{60}Co absorbed doses in water. An average value of radiation sensitivity of teeth (measured in freshly exposed samples and averaged over all multi-tooth samples) was used to convert the OSL intensities into units of absorbed dose. Fading was accounted for using the coefficients available from Sholom et al., 2011.

3. Results and discussion

The results of dose reconstruction using EPR and OSL techniques are shown in Table 1. EPR doses are given for multi-tooth samples (i.e. were averaged over six teeth of each of five multi-tooth samples) for comparison with corresponding OSL doses. OSL doses are given before and after correction on the fading of OSL signals. It is seen that EPR doses are very close to 5 Gy with an average value of 5.02 Gy and an uncertainty of 0.08 Gy (one standard deviation). These doses were considered as references to compare with the corresponding OSL-reconstructed values. Fading-corrected OSL doses for multi-tooth samples have an average of 5.15 Gy with the standard deviation of 0.69 Gy.

Table 1
Results of dose reconstruction for 5 multi-tooth samples using EPR and OSL techniques.

Multi-tooth sample#	EPR dose, aver ± std, Gy	OSL dose, Gy	Fading correction factor	OSL corrected dose, Gy
1	4.99 ± 0.30	1.20	0.28	4.27
2	5.05 ± 0.23	1.72	0.28	6.14
3	4.92 ± 0.28	1.34	0.28	4.80
4	5.01 ± 0.39	1.49	0.28	5.32
5	5.13 ± 0.31	1.46	0.28	5.21
Average, Gy	5.02			5.15
Std, Gy	0.08			0.69

Minimum measurable doses were determined at post-irradiation times of 10 min and 1 day using the same algorithm as described in Sholom et al., 2011 and were in the range 0.14–0.25 and 0.9–1.5 Gy, correspondingly (Table 2). These values satisfy the requirements for emergency triage dosimetry (Sullivan et al., 2013); a further increase in sensitivity of the OSL technique is expected if more teeth from the same individual are available for signal accumulation.

The EPR sensitivity of teeth (EPR signal normalized to the sample mass and the dose value) was compared with the corresponding OSL sensitivity (OSL signal normalized to the sample surface and the dose value) for the same teeth; no correlation was observed. This may suggest that the tooth enamel centers measured by the EPR and OSL techniques are different.

Then time stability of the EPR and OSL signals were compared for the samples derived from the same teeth. Examples of EPR and OSL signal evolution with time are shown in Fig. 1. A strong reduction is observed for the OSL signal intensity while the most prominent radiation-induced EPR signal (attributed mainly to CO_2^- radicals) remains equivalent.

The most significant changes in the EPR spectra with time after irradiation were observed in the low-field (left) region; these signals do not contribute to the dosimetric measurement. This spectral region was studied in more detail to search for any correlation with the observed properties of OSL signals. First, a check of changes in the EPR spectra with microwave power found that the signal around $g = 2.0115$ increased significantly with power (see Fig. 2 where microwave power is changed from 0.025 mW for the top spectrum to 200 mW for the bottom one). Signals in this spectral region have been attributed to CO_3^- radicals (Fattibene and Callens, 2010) that decay quickly after exposure (see Fig. 3 where shown are

Table 2
Values of MMD for multi-tooth samples measured at different times after exposure.

Multi-tooth sample#	MMD-10 min, Gy	MMD-1 day, Gy
1	0.19	1.16
2	0.21	0.85
3	0.24	0.9
4	0.25	1.48
5	0.14	1.14
Average, Gy	0.21	1.11

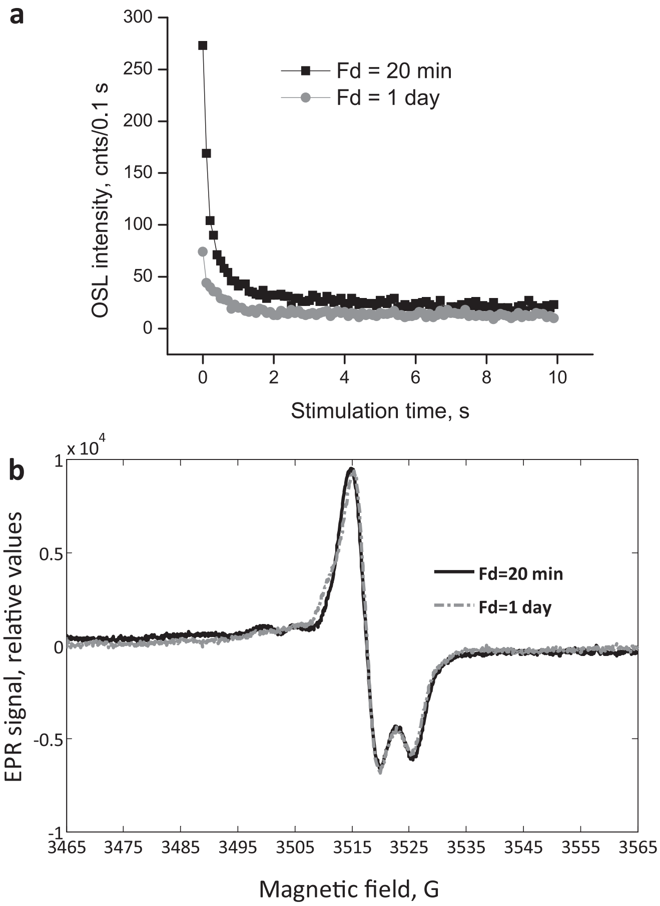

Fig. 1. Evolution of OSL (a) and EPR (b) signals with time after exposure.

EPR spectra recorded at different times after sample irradiation to 30 Gy). Calculations of the OSL and EPR signals measured at 20 min and 24 h after exposure are presented in Table 3. As it is seen from these data, both the OSL and EPR signals lost about 70% of their intensities over the 24 h period. These measurements are good

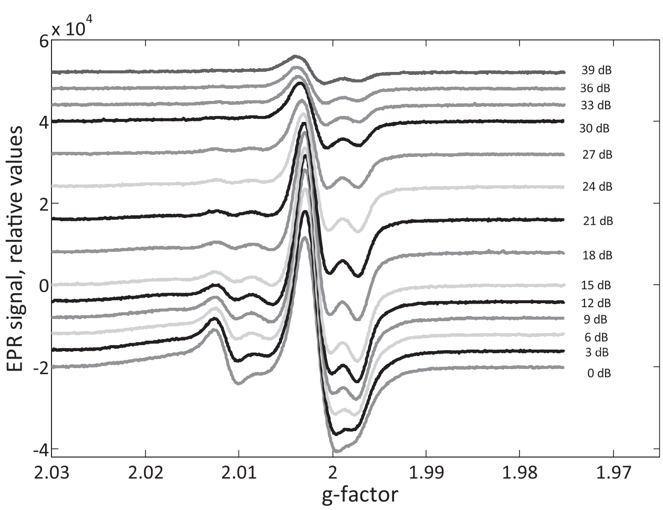

Fig. 2. Microwave power dependence of radiation-induced EPR signals in tooth enamel. "0 dB" corresponds to the maximum value of microwave power (about 200 mW). A prominent EPR signal around $g = 2.0115$ is attributed to CO_3^- radicals.

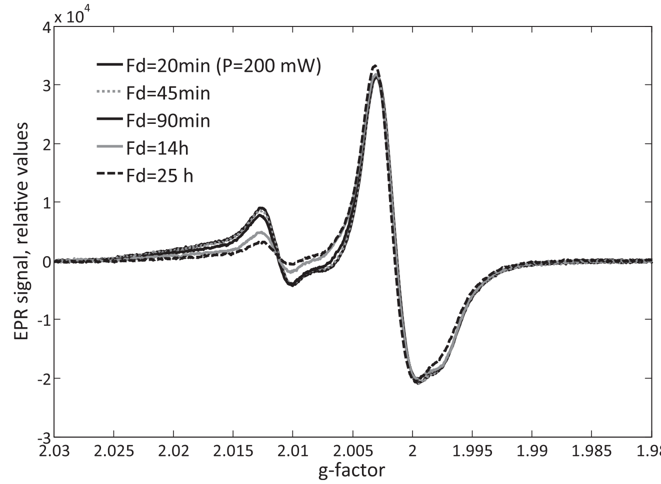

Fig. 3. Fading of the EPR signal at $g = 2.0115$. A sample was exposed to 30 Gy and recorded at microwave power of 200 mW. Values of fading times (Fd) are given in the legend.

Table 3
Comparison of OSL and EPR (at $g = 2.0115$) signals in a tooth enamel sample.

Time after exposure	OSL signal intensity	EPR signal intensity (at $g = 2.0115$)
20 min	752	502,018
24 h	220	149,845
Signal ratio	0.29	0.30

evidence for the EPR center at $g = 2.0115$ being equivalent to the OSL center used for dosimetry.

4. Conclusion

OSL reconstructed doses were found to be in agreement with corresponding EPR doses for the same teeth. Minimum measurable doses for the OSL technique were estimated to be in the range 0.9–1.5 Gy for the measurement time one day after exposure. These values satisfy the requirements for emergency triage dosimetry. A further increase in sensitivity of the OSL technique is expected if more teeth from the same individual will be used for signal accumulation.

The fading of OSL signals from teeth correlated with fading of radiation-induced EPR signals observed at $g = 2.0115$ that are attributed to CO_3^- radicals, suggesting they may be of common origin.

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