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Radiat Meas. Author manuscript; available in PMC 2012 September 1.

Published in final edited form as:

Radiat Meas. 2011 September 1; 46(9): 778–782. doi:10.1016/j.radmeas.2011.03.008.

Emergency Dose Estimation Using Optically Stimulated Luminescence from Human Tooth Enamel

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Abstract

Human teeth were studied for potential use as emergency Optically Stimulated Luminescence (OSL) dosimeters. By using multiple-teeth samples in combination with a custom-built sensitive OSL reader, ^{60}Co-equivalent doses below 0.64 Gy were measured immediately after exposure with the lowest value being 27 mGy for the most sensitive sample. The variability of OSL sensitivity, from individual to individual using multiple-teeth samples, was determined to be 53%. X-ray and beta exposure were found to produce OSL curves with the same shape that differed from those due to ultraviolet (UV) exposure; as a result, correlation was observed between OSL signals after X-ray and beta exposure and was absent if compared to OSL signals after UV exposure. Fading of the OSL signal was "typical" for most teeth with just a few of incisors showing atypical behavior. Typical fading dependences were described by a bi-exponential decay function with "fast" (decay time around of 12 min) and "slow" (decay time about 14 h) components. OSL detection limits, based on the techniques developed to-date, were found to be satisfactory from the point-of-view of medical triage requirements if conducted within 24 hours of the exposure.

Keywords

luminescence; emergency dosimetry; OSL; teeth; fading; radiation sensitivity

1. Introduction

Despite increasing needs in reliable emergency biodosimetry and promising first results demonstrated by optically stimulated luminescence (OSL) dosimetry using human teeth, the possibilities of teeth as in-vivo OSL dosimeters remain incompletely studied. Since the first publication on this topic (Godfrey-Smith & Pass, 1997) only a few investigations have been conducted and published (Pass et al., 2003; Yukihara et al., 2007; Godfrey-Smith, 2008; DeWitt et al., 2010). Summarizing the information available therein, the following important aspects of OSL dosimetry with teeth should be noted. Significant progress has been achieved with respect to the improving the minimum measurable dose (MMD): from 15 Gy in Godfrey-Smith and Pass (1997) to 1.4 Gy in Godfrey-Smith (2008) and 1–5 Gy in DeWitt

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et al. (2010). It should be mentioned here that the term “minimum measurable dose” is used by DeWitt et al. (2010); Godfrey-Smith (2008) exploits a term “minimum dose detection” and Yukihara et al. (2007) prefer a term “minimum detectable dose”. Despite this apparent difference, all terms have the same meaning: they define a dose derived from an integrated radiation-induced OSL signal equal to the corresponding background OSL signal plus three times the standard deviation of the background signal. We will follow this definition when use the term “MMD”.

A cursory examination of the achieved MMD values might suggest that the use of OSL dosimetry using teeth is satisfactory for post-exposure triage where a sensitivity (MMD) of a 2 Gy is required (Rea et al., 2010). However, the achieved values of MMD to-date were obtained in laboratory conditions on samples measured immediately after exposure. In actual emergency conditions, it is impossible to ensure that measurements of potentially exposed people can be conducted immediately after irradiation. For this reason, fading of the OSL signals from teeth needs to be taken into account. According to DeWitt et al. (2010), fading of the OSL signal from teeth was observed to be in the range 40–65% over the first 12 hours following radiation exposure. This diminution of the signal would correspond to an increase in the MMD by a factor of approximately two if the OSL measurement were to be conducted at that time. At longer times after exposure, the available data on fading characteristics contradict one another: complete disappearance of the OSL signal 2 days after exposure was predicted by Godfrey-Smith (2008) whereas DeWitt et al. (2010) noted that the OSL signals were reduced by 40–75% for the same fading time but were still observed at 40 days after exposure. Additional experiments are required with respect to both improvement of the sensitivity of the OSL dosimetry technique and greater definition of the characteristics of fading.

An additional important aspect of emergency dosimetry using OSL from teeth is the calibration of the OSL signals in units of absorbed dose. The commonly used calibration procedure of irradiating samples in the laboratory using a known dose of radiation, such as used in studies of electron paramagnetic resonance (IAEA 2002), cannot be in in-vivo applications. As is obvious, it will not be possible to irradiate each tooth in vivo to obtain the proper calibration. Therefore, DeWitt et al. (2010) proposed using a “universal average” value of the radiation sensitivity to derive a calibration factor that could be applied to all samples. The approach was tested by DeWitt et al. (2010) using a few tens of teeth and the authors demonstrated variations greater than 100%. It should be noted, however, that only small tooth fragments were used by DeWitt et al. (2010) and they demonstrated a large inhomogeneity of the radiation-induced signal even within a single tooth. One might assume that results would be different if the radiation sensitivity were to be measured using whole teeth (either buccal or lingual sides) or using a few teeth from the same individual. In the latter case, it should be possible to define an average radiation sensitivity for teeth from a single individual (individual’s radiation sensitivity) and to possibly use that value (averaged over many individuals) for calibration purposes. This modified approach is of interest for potential in vivo OSL dosimetry techniques wherein the OSL signals would be measured and calibrated using several teeth from the same individual.

An alternative approach proposed by DeWitt et al (2010) was to use UV light for normalization of the OSL signal. This suggestion implicitly assumes that the same OSL centers are formed in the tooth enamel by both the ionizing and UV irradiation and that the ratio of ionizing radiation sensitivity to UV sensitivity is a constant. In this case, the constant may be considered as a characteristic of teeth and could be determined by experiment. To investigate the relationships of sensitivity to ionizing and UV radiation, a study was designed to include: (a) readout of OSL signals following the “emergency event” exposure; (b) additional exposure of the sample to a certain test UV dose; (c) readout of the UV-

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induced OSL signal; (d) normalization of the sample's sensitivity by dividing the OSL signal from the exposure event by the OSL signal from the standardized UV exposure.

The goal of the present study was to investigate the OSL properties of teeth with respect to the above-mentioned aspects. The following tasks were prioritized:

- Improvement in the sensitivity of the OSL dosimetry technique;
- quantifying the variability of the radiation sensitivity for different teeth and different individuals;
- comparison of OSL signals due to beta and UV irradiation;
- quantifying the degree of fading of the OSL signal;

and

- determination of the MMD under a variety of conditions and comparison with the triage dose value (2 Gy was used as a cutoff triage dose that separates minimal-care and variable-care triage categories, see Rea et al., 2010).

The long-term goal of our research is to develop an emergency in-vivo dosimetry technique, using human teeth as a biodosimetry material to be applied for purposes of medical triage.

2. Materials and Methods

Information about the samples we studied is given in Table 1; examples of multiple-teeth samples are shown in Figure 1. Both large-area slices and powdered grains were used. A custom-designed OSL reader was constructed to record OSL data from samples of large size. High-power Luxeon V Star LEDs of different wavelengths were used as sources of stimulation light. Emission from the teeth was registered in the UV range by a 9235QA ET Enterprises photomultiplier tube (PMT) and a photon counter. The parameters of the OSL reader were optimized with respect to maximizing the signal-to-noise ratio. Thus, the OSL reader uses cyan LEDs (490–520 nm) as a source of stimulation light, with GG435 Long Wave Pass filters for separation of stimulation and emission light. The photon counter discrimination level was set at 10 mV, used in conjunction with a low-noise photomultiplier tube operated at 1275 V. The counting time for each data point was 100 ms and the overall stimulation time for each OSL curve was between 50 and 100 s. The dosimetric signal (DS) was defined by integrating under the OSL curve over the first 1-s interval.

Teeth were exposed with a $^{90}Sr/^{90}Y$ 250 mCi beta source to study the response to ionizing radiation. This source was calibrated against a $^{90}Sr/^{90}Y$ beta source incorporated into the Riso TL/OSL reader using thin Al_2O_3:C dosimeters. The latter source was calibrated, using similar Al_2O_3:C dosimeters, against a NIST ^{60}Co secondary standard source that itself was calibrated by NIST in terms of absorbed dose to water. Therefore, all doses quoted in this paper refer to absorbed dose to water using the NIST ^{60}Co source. The dose rate of 250 mCi source was estimated to be approximately 0.2 Gy/s at the distance about 3 cm from the source. If lower dose rates were required, one or two 1-mm aluminum sleeves were placed between the source and the sample. Homogeneity of dose deposition within working area (a circle of 18 mm in diameter) was also measured with Al_2O_3:C dosimeters and was within 7%. Five teeth were also irradiated with an X-ray source (120 kVp, 1.7 mm Al filtration) to check the possible similarity/difference between beta- and photon-induced OSL signals. UV sources emitting at 254 nm, 302 nm and 365 nm were used to study the response to UV. Two different procedures were used to analyze the OSL data: integration of the OSL curves over specific time intervals; and fitting of the OSL curves by different analytical functions, e.g., single-, bi- and triple-exponential functions. Examples of the application of these procedures are shown in Figure 2. Figure 2**a** shows how the dosimetric signal (DS) was

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obtained from the curve by integrating over the first 1 second (10 data points) and subtracting the mean of the background level. The background was determined as an average OSL signal for the last 5 s of OSL stimulation multiplied by ten (number of data points); noise was defined as the standard deviation of the background multiplied by the square root of ten. The minimum measurable dose was determined as that dose for which DS is background plus three times the standard deviation of the background. Figure 2**b** shows example results for fitting the decay curve to a bi-exponential, plus constant background. This method was useful for comparison of the shapes of the OSL curves.

3. Results and Discussion

3.1 Sensitivity and dose-response curve

Using the measurement system described, OSL curves were recorded for different samples exposed to different doses. At the beginning, beta- and X-ray-induced OSL signals were compared for 5 samples exposed to the test dose of 10 Gy. Within experimental errors, OSL signals were found to be the same for the same samples (but varied significantly between different samples). This finding could be explained by the fact that UV light attenuates strongly with depth in teeth (Sholom et al., 2010); as a result, most of the radiation-induced OSL signal collected by PMT comes from thin surface layer of an exposed sample (with thickness of about 0.1 mm). Taking into account that attenuation of both beta- and X-ray radiations is small enough within 0.1 mm range (about 10 % for a $^{90}Sr/^{90}Y$ beta radiation according to Yang et al. (1998) and much smaller for X-rays used, see e.g. Sholom et al. (2007)), one can expect approximately the same energy deposition for the surface 0.1 mm layer of a tooth from both types of radiation for the same values of entrance dose. So, a $^{90}Sr/^{90}Y$ beta source was used as a substitute of a photon source in all experiments described below.

The OSL curves were analyzed using the procedure illustrated in Figure 2**a** and the corresponding values of DS, background, noise and MMD were determined. The dose response curves were observed to be linear for all tested samples for doses below 10 Gy, the dose range of interest in triage applications. An example of a dose-response curve is shown in Figure 3 for a representative multiple-teeth sample studied immediately after exposure; the MMD is 27 mGy. The MMDs for other tested multiple-teeth samples were in the range 0.03–0.64 Gy, lower than the values reported by Godfrey-Smith (2008) and DeWitt et al. (2010) by about one order of magnitude. This significant improvement in sensitivity was achieved through the use of a higher power of light stimulation (power at the sample location is estimated to be 500 mW cm^{-2}) and through the use of a larger sample surface for stimulation and emission of the OSL. Typically, the net surface of the multiple-teeth samples was in the range 2–2.5 cm^2. The use of a higher stimulation power resulted in the near depletion of the OSL signal during just 1 s of stimulation. Stimulation over several tens of seconds was required in the work of Godfrey-Smith (2008) and 5–10 s in the work of DeWitt et al. (2010). These findings suggest that by using high-power LEDs and larger area samples, the time for OSL signal integration (1 s in the current case) and, hence, the noise integrated over this same time period, can be reduced. Here, it should be noted that both the signal and the noise increase when the number of teeth, N, used in the multiple-teeth sample is increased. While the OSL signal increases proportionally to N (on average), the noise increases proportionally to $N^{1/2}$. As a result, the signal-to-noise ratio increases proportionally to $N^{1/2}$. It should also be emphasized that most of the multiple-teeth samples used in the present work consisted of 3–4 teeth and, therefore, further improvement of sensitivity is expected by using more teeth from each individual for each measurement. The improvement in sensitivity that can be achieved by increasing the number of teeth measured will be a clear advantage for in-vivo measurements.

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3.2 Variability of radiation sensitivity

The grained samples were measured in a single-layer geometry. The volume sensitivity (i.e., the OSL signal was normalized to the sample weight) of the single-layer of 630–850 μm diameter samples was approximately two times smaller than for grains of 250–425 μm. This observation is consistent with a surface character for the OSL phenomenon in teeth. For this reason, radiation sensitivity in successive experiments was determined using surface normalization only.

Intra-person variability of radiation sensitivity (i.e., sensitivity between different teeth of the same person) was measured using 8 pairs of front teeth and 20 trios/quartets of molars and was found to be within 11–39% for incisors and 13–139% for molars. Inter-person variability was determined for both single-tooth and multiple-teeth samples (i.e., for teeth and for individuals) and was found to be 115% and 53%, respectively. It can be deduced that while individual calibration is needed for high-precision estimates, for cases when 50% accuracy in the estimated radiation dose is sufficient, a universal calibration curve derived from multiple-teeth samples might be applied.

3.3 Comparison of OSL signals due to beta and UV irradiation

In general, no correlation was found between OSL signals obtained after exposure to beta with those after UV radiation. To explain this phenomenon, it was hypothesized that beta and UV radiation activate different OSL centers in teeth. This was supported by fitting the corresponding OSL curves with bi-exponential decay functions. An example of the OSL curves following beta and UV (302 nm and 254 nm) irradiation is shown in Figure 4. Different curve shapes were observed and fitting the data resulted in the decay time constants of 0.14 and 3.2 s for beta and 1 and 16 s for both 302 and 254 nm UV OSL. Thus, we conclude that although 302 and 254 nm UV activate the same OSL centers, these are different from those activated by beta radiation. This finding makes it impossible to use 302 and 254 nm UV for calibration of radiation-induced OSL signals in teeth, but leaves open the possibility of using other wavelengths for such an application.

3.4 Fading

For samples that were stored under routine laboratory illumination following radiation exposure, the DS signal diminished by 50% during the first 5 minutes and by 80% during the first hour. For samples stored in the dark, it was observed that the teeth could be divided into one of two groups: one group characterized as displaying typical fading in which the DS monotonically decreased with time after exposure, while the other group displays had atypical fading characteristics in which the DS changed non-monotonically with time after exposure. The latter group, in some cases, could display one or several local maxima. Ninety-eight percent of tested samples were in the first category since they demonstrated typical fading; only 3 samples (all incisors) were in the second category and displayed unusual fading. It was observed that samples with the different fading characteristics could be distinguished by the shape of the corresponding OSL curves. Fitting the OSL curves with a bi-exponential decay function, all samples displaying unusual fading were characterized by much higher values for the decay time constants compared to samples with typical fading properties, especially for the faster bleaching component. For example, the average value for the fast component in the samples with atypical fading characteristics was 0.6±0.06 (s) compared to 0.16±0.04 (s) for the samples with more typical fading characteristics.

Typical fading dependencies could be fitted with a bi-exponential decay function with decay time constants of 0.2 and 14 h (see Figure 5). Our data for teeth with typical fading properties indicate that approximately 20% of the initial signal is left after 1 day and 5% after 3 weeks following radiation exposure. These findings are in qualitative agreement with

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data obtained by DeWitt et al. (2010). Comparison with the data of Godfrey-Smith (2008), however, requires some comment. In fact, the fading data published in Godfrey-Smith (2008) were collected only for fading times of less than 10,000 s (about 3 h) and then extrapolated for longer time intervals using a single logarithmic decay function (i.e. a single exponential decay function on a linear scale). However, our results demonstrated that there are at least two different components in the fading curve and fitting with a single exponential decay function is not possible when fitted over a wide interval of fading time. If we compare the raw experimental data from the two works, however, the coincidence is good. For example, for a fading time of 3 h the value of fading was about 54 % in Godfrey-Smith (2008) and about 57 % in our work.

4. Extrapolation to triage situations

Since our goal is to develop a method suitable for dosimetry for individuals following an actual exposure situation we have estimated how the MMD would change if we used variable numbers of teeth, ranging from 4 teeth in one individual to 16 teeth in one individual (all molars and premolars of an individual excluding wisdom teeth). These estimates assume the same efficiency for detecting OSL as for the current measurements and similar sensitivities per tooth as were found in the present work. Further, we assumed that the OSL measurements were conducted 24 h after the radiation exposure. To get the most accurate estimation, we used experimental values of the MMD measured for 20 multiple teeth samples 24 h after laboratory exposure. Estimations of the MMD for 16 teeth measurements were obtained by dividing the initial values of MMD by a coefficient being equal to square root of the ratio of 16 to N, where N is the number of teeth actually used in the experimental multiple-teeth samples. Results of calculated MMD are shown in Figure 6 together with corresponding initial values of MMD. Based on the data on OSL sensitivity obtained to-date, our analysis suggests an estimated 80% probability of detecting a dose of 2 Gy when using 16 teeth in the measurement conducted 24 h after exposure. Further, we estimate that the probability of detecting a dose of 2 Gy would increase to 95% if the measurements were conducted 12 h after the radiation exposure.

5. Conclusions

With the custom-designed OSL reader used for this work, and using multiple-teeth samples, we determined that a minimum measurable dose of less than 0.64 Gy is feasible under a range of conditions. An experimental minimum value of 27 mGy was obtained for the most sensitive sample for OSL measurements made immediately after exposure. The variability of the OSL sensitivity is 115% for teeth and 53% for individuals (when multiple-teeth samples are used).

The OSL detection limits, based on the techniques developed to-date (using multiple teeth) are satisfactory from the point of view of medical triage requirements if the OSL measurements are conducted within 24 hours of the exposure.

The results accumulated to-date were obtained using laboratory measurements of extracted teeth. In vivo measurement technique yet to be developed and studied.

Taking into account the very short times for the OSL measurement (seconds to minutes), the relatively simple requirements for OSL equipment (which can be made field deployable and transportable by a single person), we conclude that OSL from teeth is a viable emergency dosimetric technique that could play a role in supplementing other technologies available, most of which are much harder to implement and require much more sophisticated equipment. Future work will attempt to study tooth surface characteristics using electron

microscopy and to investigate possible correlations of sensitivity with age, ethnicity, and other variables.

Acknowledgments

This work was supported by the Intra-Agency agreement between the National Institute of Allergy and Infectious Diseases and the National Cancer Institute, NIAID agreement #Y2-Al-5077 and NCI agreement #Y3-CO-5117.

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Figure 1.
General appearance of the multiple-teeth samples used in the present study (the scale is in cm).

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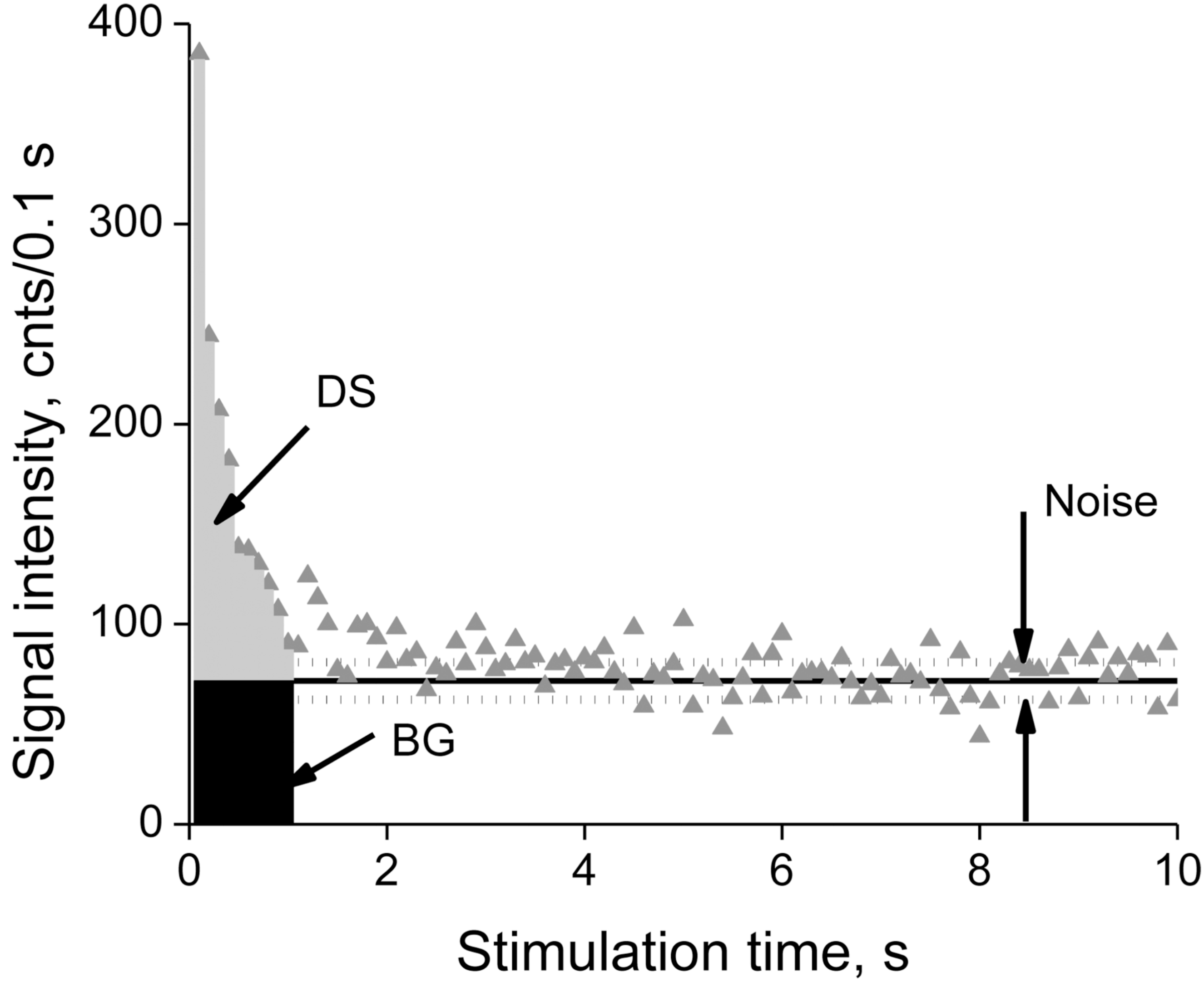

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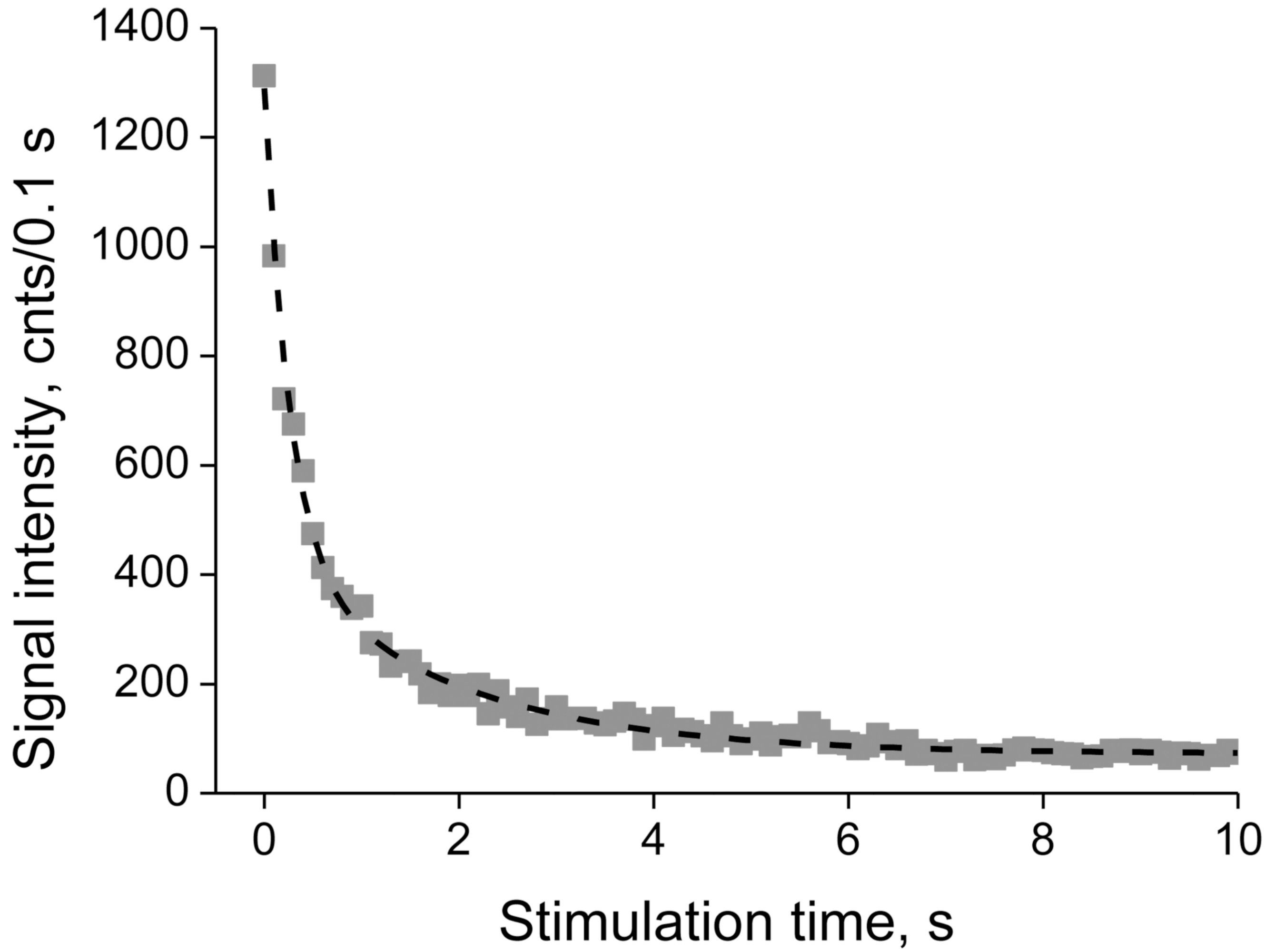

2b

Figure 2.

Illustration of two algorithms developed for OSL signal analysis. **a** - integration of OSL curves for different time intervals (1 s in given example); **b** – fitting of OSL curves using a bi-exponential decay function. For the sample shown in plot **a**, beta dose was 2 Gy; the ratio of DS and noise was 35 and MMD was 170 mGy. For the sample in plot **b**, the best fit was observed with a bi-exponential decay function: y = y0 + A1*exp(−x/t1) + A2*exp(−x/t2); bleaching time constants were 0.26 and 1.87 s.

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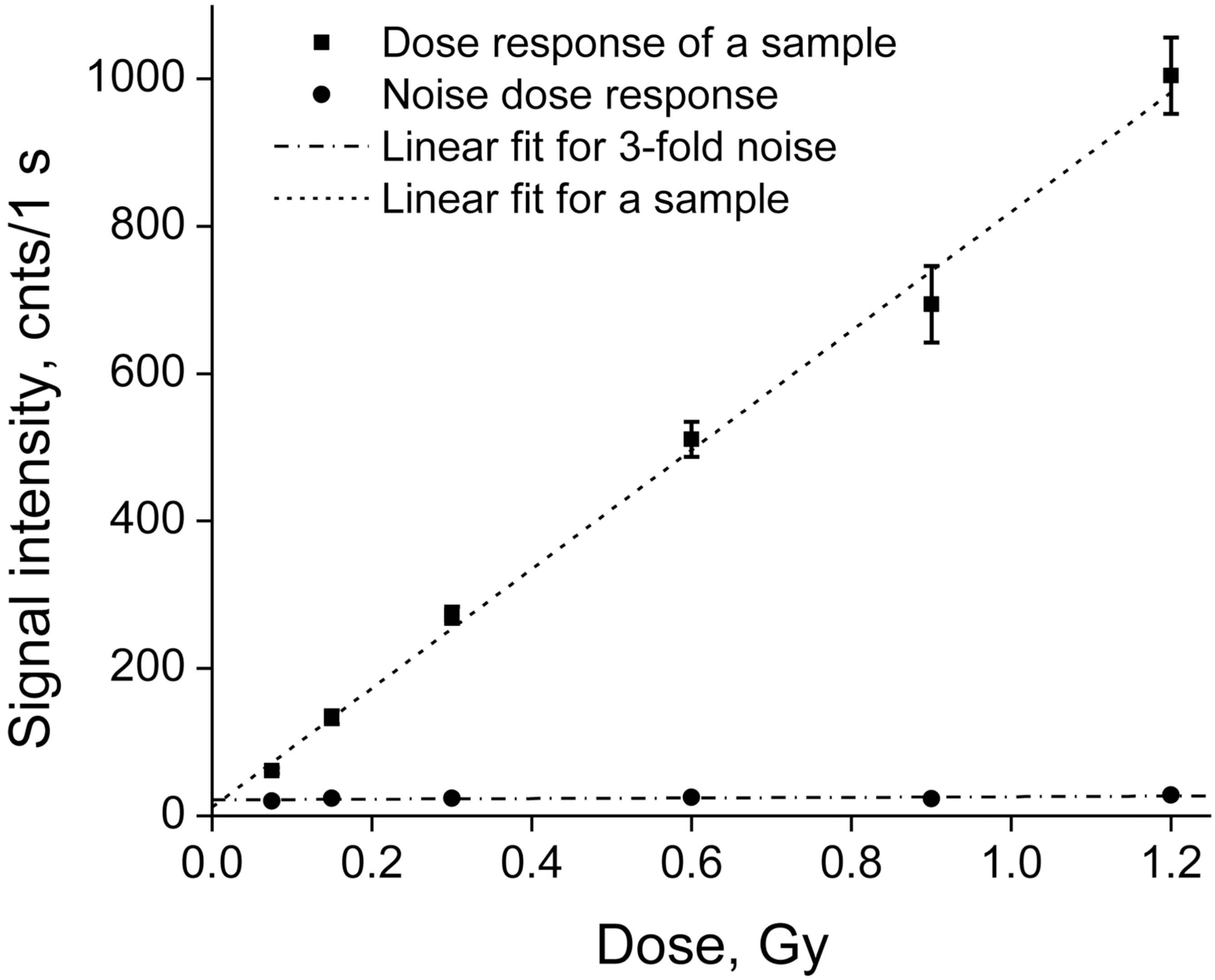

Figure 3.

Dose response curve for a representative multiple-teeth sample studied immediately after exposure. The MMD is 27 mGy.

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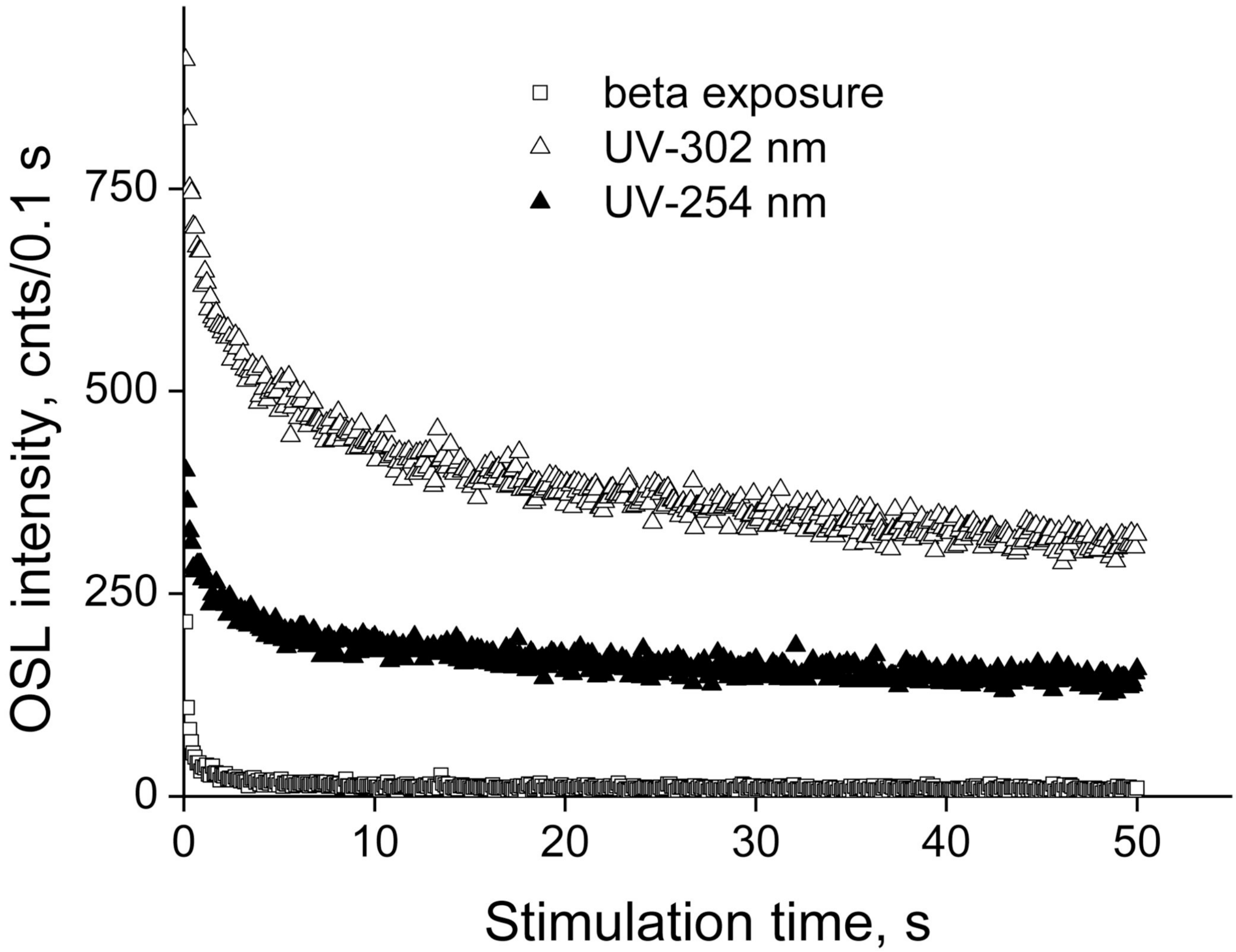

Figure 4.

Example OSL curves following beta and UV (302 nm and 254 nm) irradiation. Values of the decay time constants obtained from bi-exponential fitting of the OSL curves are: 0.14 and 3.2 s for beta OSL; 1 and 16 s for both 302 and 254 nm UV OSL.

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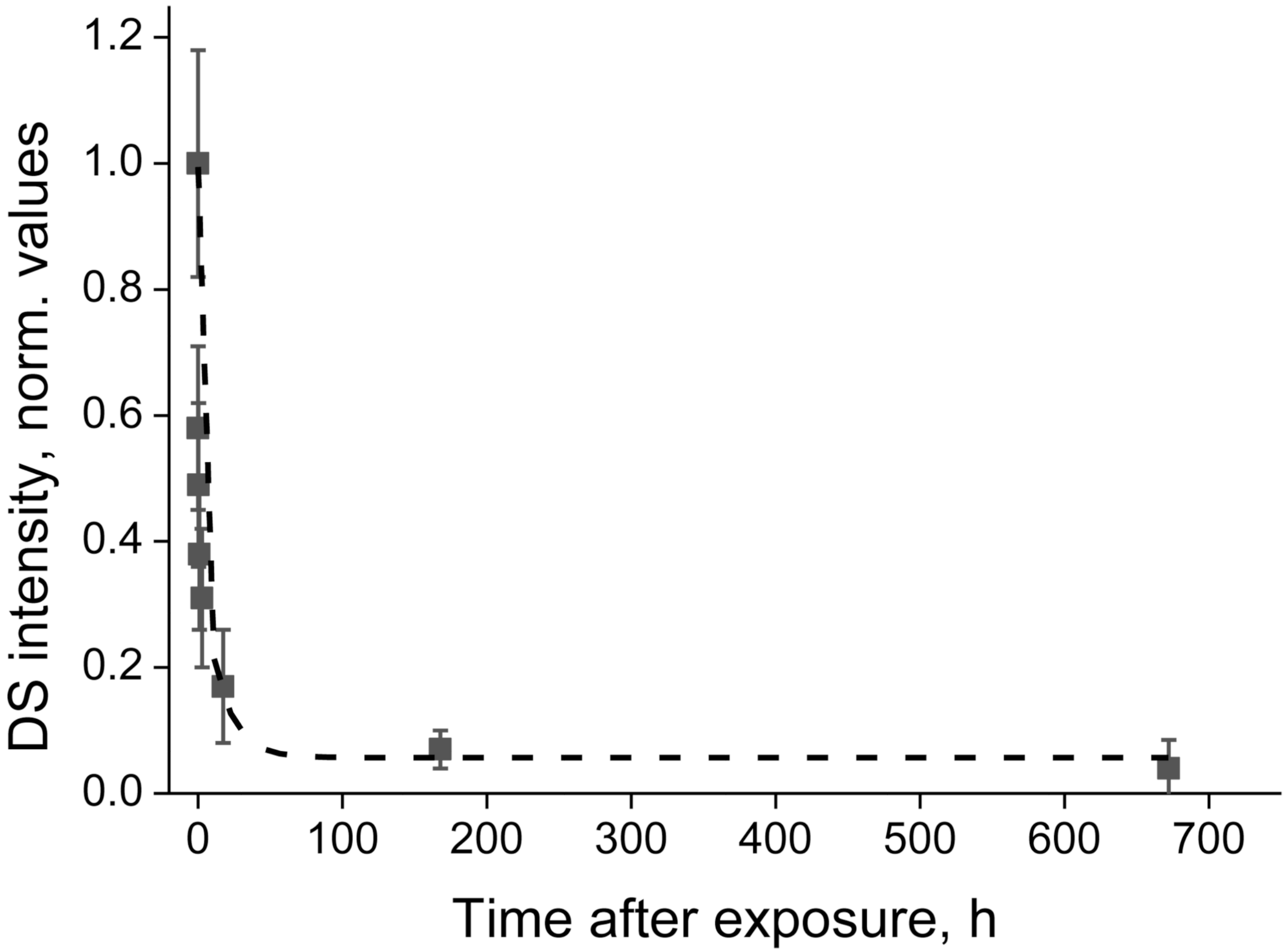

Figure 5.
Fading of the DS in samples with "typical" fading properties. Every point is an average of measurement of 18 different teeth; the error bars show the standard deviations. All data were normalized to the corresponding values of DS measured in samples immediately after exposure. The best fitting was with a function y = y0 + A1*exp(−x/t1) + A2*exp(−x/t2); values of decay time constants were 0.19 h and 14 h.

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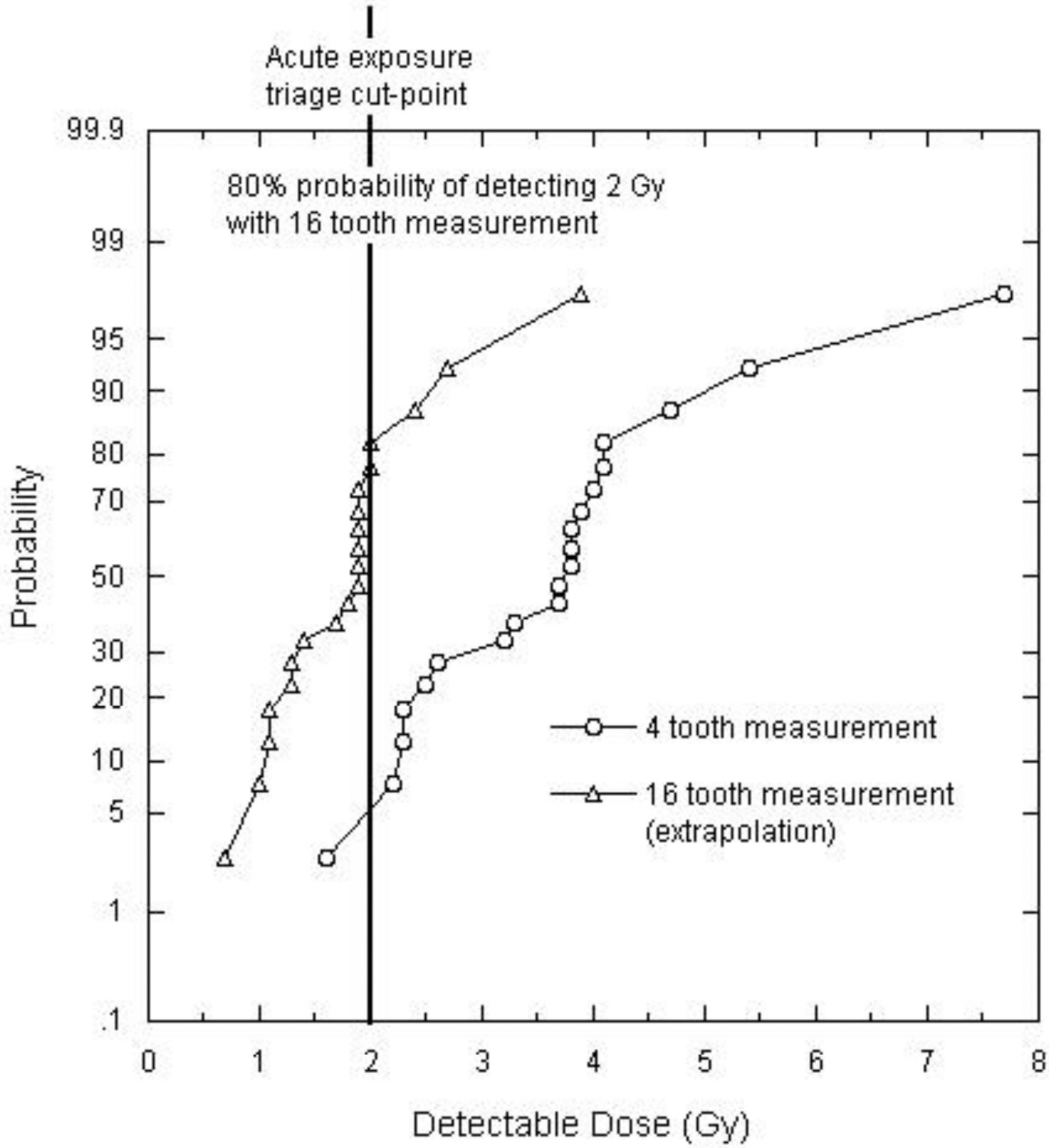

Figure 6.
Empirical cumulative probability of minimum measurable dose for 4 teeth (open circles) or 16 teeth (open triangles) from a single person at 24 hours after exposure.

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Table 1

General description of teeth and tooth samples used in the study

Teeth source and quantity	Samples prepared and tested
100 teeth extracted for medical reasons at the NIH from 35 individuals with available information about age, gender, tooth position (all molars)	• Simulators of whole teeth (single-tooth samples): enamel layers with thickness of 2–3 mm cut from tooth buccal or lingual side • Multi-teeth samples: single-tooth samples from three and more teeth of the same person measured together (Figure 1)
50+ teeth extracted for medical reasons at local dental clinics without any individual information (impersonal teeth, both molars and incisors)	• Grained samples of different size (250–425 μm, 425–630 μm and 630–850 μm) studied in single-layer geometry • Simulators of whole teeth: buccal halves of incisors and 2–3 mm enamel layers cut from molars • Multi-teeth samples