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Comparison of γ- and UV-light-induced EPR spectra of enamel from deciduous molar teeth

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Abstract

From previous work, it is known that CO_2^- radicals in tooth enamel are induced by gamma as well as by UV-light exposure. The parameters of the EPR signal of the CO_2^- radical were found to be independent of the source of exposure. However, it would be desirable for retrospective dosimetry to identify other characteristic features of the EPR spectrum of tooth enamel, which would allow differentiation between the two sources of exposure. In the present work, enamel of deciduous molars was exposed to γ-radiation from a ^{60}Co-source and 254 nm UV-light from a low-pressure mercury lamp. The resulting EPR spectra were deconvoluted, and the native spectrum simulated from spectra of the CO_2^- radical, and two further EPR lines. Both EPR signals of the native spectrum were located at $g = 2.0046$, but were different in line shape and width. One was a 1 mT wide isotropic signal of Gaussian line shape while the other was a 0.7 mT wide axial signal of Lorentzian line shape. A comparable study of the amplitudes of the native and CO_2^- signals was done before and after γ- and UV-light exposure. While the native signals were found to be only slightly sensitive to γ-radiation, their amplitude increased significantly on UV-light exposure. Feasibilities are discussed to distinguish different radiation sources by exposure-induced alterations of the native EPR spectrum.

Keywords: EPR; Tooth enamel; Deciduous teeth; UV-light

1. Introduction

During the last two decades, electron paramagnetic resonance (EPR) dosimetry using tooth enamel has become a subject of interest. This is due to its great potential in dose reconstruction for accidents and epidemiology e.g. for the atomic bomb survivors of Hiroshima and Nagasaki (Ikeya et al., 1984) and the nuclear workers of Mayak (Romanyukha et al., 1994; Wieser et al., 1994; Tolstykh et al., 2000). EPR-enamel-dosimetry is based on the detection of radiation-induced CO_2^- free radicals in hydroxyapatite (HA) microcrystals [$Ca_{10}(PO_4)_6(OH)_2$]. During the mineralization process of HA, phosphate and/or hydroxyl ions are substituted by carbonate ions. These carbonate ions become CO_2^- radicals upon absorption of ionizing radiation. With other signals that share the signal of CO_2^- these compose the EPR spectrum of the tooth enamel. The sources of such other signals are not known at present but they are possibly due to organic and other impurities, or defects in the crystal structure of HA. The native signals heavily obscure the CO_2^- signal for absorbed doses below about 500 mGy. The intensity of the native signals can be minimized by appropriate sample preparation procedures (e.g. Polyakov et al., 1995; Ivannikov et al., 2001)

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and can be discriminated by spectral deconvolution (e.g. Hayes et al., 1997; Koshta et al., 2000).

The analysis of reconstructed doses from teeth at different tooth positions revealed a large variation of results from incisors versus molars, as well as a large variation of results from the buccal versus lingual part of incisors (Ivannikov et al., 1997). These differences most probably result from UV exposures. It is well known that the sun has three UV components: UVC (100–280 nm), UVB (280–315 nm) and UVA (315–400 nm). The atmosphere absorbs the former, while the other two types reach the surface of earth and might induce free radicals in tooth enamel (Liidja et al., 1996). Other possible sources of UV exposure are solariums UVA light and UVA/B-light emitted in course of dental treatment.

Since there is no distinction between the induced free radicals from different sources of radiation (γ-, X-rays or UV), it would be important to have a manner of identifying the different sources of exposures. To achieve this task, in the present work the intensity of EPR signals of deciduous molar teeth in dependence on UV-light and ^{60}Co-γ-ray exposure were compared.

2. Materials and methods

The present study was done with 4 deciduous molars from 2 children of the same age, from Munich, Germany. The teeth were removed naturally. All samples were washed with 0.1 M Titriplex solution for one-quarter hour in an ultrasonic bath at room temperature. EPR measurements were taken after which the black spots and blood remnants were removed neatly; teeth were washed with ethanol and dried under vacuum for 2 h at 40 °C. The samples were placed in quartz tube with an internal diameter of 2.5 mm. The EPR spectra were recorded at room temperature with a Bruker ECS 106 spectrometer operating in X-band. The experimental parameters were as follows: microwave power 25 mW, time constant 163 ms, magnetic field sweep 5 mT, modulation amplitude 0.145 mT, conversion time 81.92 ms and number of accumulations 40.

After measurement of native signals, the samples were divided into two groups. Three samples were irradiated to various γ-doses (0.1, 0.5, 1 and 10 Gy) using a ^{60}Co source (Type Eldorado) with a dose rate 50 mGy/min at the outer surface of phantom box of 0.5 cm thick walls at all sides. The inside space of the phantom box was 1 cm in the direction of the beam and $9 \times 9\,cm^2$ perpendicular to the beam. The distance from the phantom to the ^{60}Co source was 100 cm and the radiation field had a size of $12 \times 12\,cm^2$. The fourth sample was exposed from the side of the enamel surface for different time intervals (15, 30, 60, 120, 240, 400 and 4210 min) to artificial UV-light using UV-low-pressure mercury lamp, type NK6/12 (Heraeus, Germany) and of power $8\,mW/cm^2$. The lamp was mounted on a stand 10 cm above the table. The stability of UV-induced signals was studied for time intervals up to 2 months.

In order to avoid the residual anisotropy arising from the assymmetrical surface of these massive deciduous samples and variations in the sample positioning in the microwave cavity, each sample was recorded five times with rotation of the sample tube by 45° between the measurements. The amplitudes of the EPR signals were averaged after individual evaluation from each recorded spectrum. All measurements were done one day after the irradiation process.

3. Results and discussion

The variation of the EPR spectrum of deciduous molars with increasing artificial UV-light exposure is shown in Fig. 1a. The amplitude of the CO_2^- signal correlates well ($r = 0.999$) with the exposure time (Fig. 2a). At an UV-exposure time of 4200 min, the concentration of the correspondingly produced CO_2^- radicals is equivalent to absorbed dose of 8.7 Gy. While the g-values of the CO_2^- and background (BG) signals seem to be identical to that induced by γ-irradiation (Fig. 1b), the graphs reveal a distinguishable difference between the signal intensities. Fig. 2 shows that the EPR spectrum can be described as a composite of two native signals and the CO_2^- radical at $g_\perp = 2.0018$, $g_{//} = 1.9973$ with peak maximum at $g = 2.0032$ and line width ΔH_{pp} $(g_\perp) = 0.45$ mT. The signal intensities of the individual components were evaluated by spectrum deconvolution. The CO_2^- signal used in deconvolution was obtained as a composite of powder simulated EPR spectra of an orthorhombic and quasi-axial CO_2^- radical (Vanhaelewyn et al., 2001, 2002; Zdravkova et al., 2003). The two native, BG1 and BG2, signals are both located at $g = 2.0046$ but with different line shape and width. BG1 was a 1 mT wide isotropic signal and was simulated with Gaussian line shape. BG2 was a 0.7 mT wide axial signal of Lorentzian line shape. The signal was simulated with an axial g tensor, $g_x = g_y = 2.0055$, $g_z = 2.0021$ and a line width of 0.5 mT for all 3 components. The parameters of the g tensor were taken from Callens et al. (1998) and were proposed to be related to surface located CO_2^- radicals.

The amplitudes of the two native and CO_2^- EPR signals by exposure to artificial UV-light up to 4200 min (Fig. 2a) and γ-irradiation in the range from 100 mGy to 10 Gy (Fig. 2b) were studied and the results can be pointed out as follows:

- The amplitude of CO_2^- signal has coincident linear response for both UV- and γ-radiations (Fig. 3a). A quadratic amplitude–dose relationship has been