Radiation Measurements 81 (2015) 257–261

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Dose response and fading studies on de-proteinated tooth enamel after de-convolution using the sum of general order kinetics and a component for tunnelling recombination

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HIGHLIGHTS

- Two different de-proteinization procedures were applied to tooth enamel.
- OSL de-convolution analysis used one component of general order kinetics plus a tunnelling component.
- De-convolution procedure resulted to a fast OSL component with enhanced signal to noise ratio.
- This OSL yielded a linear dose response throughout the entire dose range and absence of anomalous fading.

ARTICLE INFO

Article history:
Available online 4 December 2014

Keywords:
OSL
Tooth enamel
Tunnelling
De-convolution
Fading
Dose response

ABSTRACT

The present study reports on the results of dose response and fading for the case of blue stimulated OSL signals from tooth enamel samples. These samples have been previously subjected to two different de-proteinization procedures, after applying hydrazine and sodium hydroxide. The OSL signal, when quantified based on the conventional way of OSL analysis, namely the use of the initial 5 s of the signal minus the intensity of the last 5 s as a background signal, yields bad signal to noise ratio as well as instability. The application of the de-convolution procedure including two components, one of general order kinetics and one of tunnelling recombination results to a fast OSL component with enhanced signal to noise ratio, isolating thus the fading component and yielding a linear dose response throughout the entire dose range of the present study. Finally, the physical meaningfulness of the fitting parameters for the tunnelling component, especially the dimensionless parameter representing the normalized donor −acceptor density ρ' is also discussed.

1. Introduction

The ever-increasing numbers of radiation-induced accidents all around the world power the retrospective dosimetry studies; one of the major challenges in these related studies is the effective application of human biological materials as dosimeters. Therefore, much effort is being put towards both *in-vivo* dose reconstruction approaches. Electron Paramagnetic Resonance (EPR) studies of nails (Trompier et al., 2014 and references there in) as well as both EPR and luminescence studies on tooth enamel (Egersdörfer et al., 1996; Godfrey-Smith and Pass, 1997; Fattibene et al., 2005, 2008; Yukihara et al., 2007; Godfrey-Smith, 2008; Güttler and Wieser, 2008; DeWitt et al., 2010; Sholom et al., 2011; Sholom and Desrosiers, 2014; Soni et al., 2014), have been recently emerged from the necessity for alternative bio-luminescent retrospective dosimeters.

Recently, Yüce et al. (2010) investigated the effect of chemical sample preparation with hydrazine reagent on tooth enamel sample sensitivity. According to their results, the blue stimulated OSL signal from enamel which has been subjected to hydrazine deproteinization process suffers from fading, while the dose

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http://dx.doi.org/10.1016/j.radmeas.2014.12.003
1350-4487/

response curve lacks of linearity. Their approach included quantification of the OSL signal based on the conventional way of OSL analysis, namely the use of the initial 5 s of the signal minus the intensity of the last 5 s as a background signal.

Tooth enamel is composed of hydroxyapatite $\{Ca_{10}(PO_4)_6(OH)_2\}$ needle crystallites, about 0.6–0.9 mm long that are dispersed in an aqueous organic gel (IAEA TEC DOC- 1331,2002). The family of apatites consists of solid-state integrating luminescent material that yields very intense TL, OSL and IRSL anomalous fading (Kitis et al., 1991; Polymeris et al., 2006; Tsirliganis et al., 2007; Polymeris et al., 2014 respectively). The anomalous fading yielded for the OSL by this material provides a strong indication towards a tunnelling mechanism being considered as a possible explanation. In this respect and in the present work, a de-convolution approach was applied to the blue stimulated OSL signals from enamel which have been subjected to hydrazine and sodium hydroxide deproteinization processes, towards discrimination of a stable OSL component.

2. Materials and methods

2.1. Samples

Adult human tooth enamel samples were collected from dental hospitals and clinics and sterilized in sodium hypochlorite solution (%5 NaOCl) for 24 h before use (IAEA, 2002). The tooth crowns were separated from the roots and the enamel layer was isolated with a diamond saw. The enamel size fractions separated from several teeth were 20μ–150μ. Tooth samples were provided by the National Institutes of Health in Turkey. Hydrazine (N_2H_4) deproteinization steps were performed as described by Ivannikov et al. (2001) except the ultrasonic bath step. Enamel samples (about 150 mg) were placed into 10 ml glass flasks, which can be sealed with covers. At the first step defatting was performed in a 1:1 mixture of ethanol with ethyl ether for 1 h. After the defatting phase, the solution was decanted and the samples were dried. These dried samples were exposed to 3 ml hydrazine for about 24 h at 55 °C in an ultrasonic bath. After this step, the reagent was decanted. Samples were subsequently washed out with 50% ethanol-water solution and with pure ethanol during one day. After the samples were dried in open air, aliquots were prepared with silicon spray and weighed.

Sodium hydroxide deproteinization steps were performed as described by Fattibene et al. (2008). Enamel samples (about 100 mg) were put in a 5 M NaOH solution for 16 h in an ultrasonic bath. After the first 8 h of treatment the samples were removed from NaOH solution and rinsed using demineralized water; the treatment in NaOH solution was then repeated for the remaining 8 h. After the samples were dried in open air, aliquots were prepared with silicon spray and weighed.

2.2. Apparatus and measurement conditions

All OSL measurements were performed with a Risø TL/OSL reader (TL/OSL-DA-20, Risø National Laboratory). The samples were stimulated in all experiments with blue LEDs (470 ± 20 nm) for 40s or 60s. All irradiations were performed after de-proteinization procedures using the built-in $^{90}Sr/^{90}Y$ beta source (40 mCi) at a calibrated dose rate of 0.141 Gy s^{-1}. Light detection was carried out with a photomultiplier tube (PMT) bialkali EMI 9235QA which has an extended UV response with maximum detection efficiency between 300 and 400 nm. To prevent scattered stimulation light from reaching the PMT, a 7.5 mm Hoya U-340 detection filter, which has a peak transmission around 340 (±80) nm, is employed (Bøtter-Jensen et al., 1999). The aliquot weight was approximately 5 ± 0.02 mg for all OSL measurements. Aliquots were prepared on stainless steel cups. All samples were stored in the dark at room temperature (20 °C) before and after irradiation.

2.3. Method of analysis

The experimentally obtained OSL curves were fitted with a sum of a tunnelling component plus a single component of general order kinetics. For the latter case the general order kinetics (GOK hereafter) expression for OSL theory (Kitis and Pagonis, 2008) was applied:

$$I = I_0 \cdot \left(1 + \left(b-1\right)\cdot\frac{t}{\tau'}\right)^{-\frac{b}{b-1}}, \quad b \neq 1 \tag{1}$$

where $I(t)$ is the intensity of the luminescence signal as a function of time, $\tau' = 1/\lambda$ (s) is the luminescence via the conduction band lifetime and b is the order of kinetics. For the case of general order kinetics the value of kinetic order b was left to vary freely in the range between 1.00001 and 2.

For the case of the tunnelling component, recombination is assumed to take place via the excited state of the donor, and nearest-neighbour recombination takes place within a random distribution of centres. Within this model, the following set of equations was applied (Kitis and Pagonis, 2013):

$$I(t) = \frac{3n_0\rho' zF(t)^2}{1 + z\,t/_{\tau}}\exp\left(-\rho' F(t)^3\right) \tag{2}$$

$$F(t) = \ln\left(1 + z\frac{t}{\tau}\right) \tag{3}$$

$$\tau = (\sigma\cdot\varphi)^{-1} \tag{4}$$

where n_0 is the initial concentration of trapped electrons, z a constant, τ the tunnelling luminescence lifetime for recombination via the excited state, ρ' a dimensionless parameter representing the normalized donor–acceptor density, σ is the photo-ionization cross section and φ the stimulation flux. The abovementioned set of equations was recently suggested by Kitis and Pagonis (2013), who have obtained analytical solutions for the set of differential equations of the model of Jain et al. (2012), by using certain mathematical and physical simplifications. These authors presented a new general kinetic model which quantifies localized electronic recombination of donor–acceptor pairs in luminescent materials. The main physical assumption in the model is the presence of a random distribution of hole traps in the luminescent volume, and an associated range of random nearest-neighbour recombination probabilities. Stimulated recombination takes place only via the excited state of the electron trap, by either optical or thermal stimulation. Similar approaches were recently adopted by Sfampa et al. (2014) for the case of prompt isothermal decay (PID) of Durango apatite as well as by Şahiner et al. (2014) for the case of IRSL at elevated temperatures of feldspar-contaminated quartz.

All curve fittings were performed using the software package Microsoft Excel, with the Solver utility (Afouxenidis et al., 2012) while the goodness of fit was tested using the Figure Of Merit (FOM) of Balian and Eddy (1977).

$$\mathrm{FOM} = \sum_i \frac{\left|Y_{\mathrm{Exper}} - Y_{\mathrm{Fit}}\right|}{A} \tag{5}$$

where Y_{Exper} is the experimental glow-curve, Y_{Fit} is the fitted glow-curve and A is the area of the fitted glow-curve. The FOM values