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RETROSPECTIVE ACCIDENT DOSIMETRY USING DENTAL CERAMICS

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Abstract. Optically Stimulated Luminescence (OSL) and Thermoluminescence (TL) properties of various brands of feldspathic ceramics employed for the fabrication of the external layer (veneer) of dental crowns have been investigated, in order to develop a dose assessment technique for medical triage following unplanned exposures of individuals to ionizing radiation. C7 dental ceramic type seemed particularly suitable for accident dosimetry purposes. It exhibited intense OSL signal, relatively high homogeneity (relative standard deviation of 13%), a linear dose response in the dose range of interest (1.5 Gy–9 Gy) and values of the minimum detectable dose ranging between 9 mGy and 300 mGy.

Key words: OSL, TL, dental ceramics, retrospective accident dosimetry.

1. INTRODUCTION

Retrospective dosimetry measures chemical, biological or physical markers of radiation exposure which persist long enough to be able to assess doses received days, weeks or years before sampling. Its main objective is to help reduce the health consequences where practicable by offering support management for the treatment of radiation injuries.

The current methods used in accident dosimetry for dose reconstruction include biological (cytogenetic, genetic, hematological, protein biomarkers), physical (electron paramagnetic resonance, luminescence dosimetry, neutron activation), and computational techniques [1]. With regard to physical methods, luminescence instrumentation is more accessible, therefore this technique has sparked great interest. The best established luminescence methods for the purposes of dose reconstruction are focused on the use of natural minerals such as quartz and feldspar that can be found in bricks, tiles and pottery collected from local buildings

[2–4]. However, in the last several years numerous studies suggested the dosimetric properties of widely used personal objects for retrospective dose assessment [5–15]. In addition to these, electronic components, memory chip modules and screens removed from personal objects, materials used for dental prosthetics restoration were also investigated aiming to determine their usefulness as retrospective dosimeters [16–19].

Dental ceramics is a term used to describe porcelain and glass-ceramic materials that are employed in the construction of crowns, restorative components of teeth and prosthetic teeth. Dental ceramics, owing to their intimate contact with the human body, are of interest as a luminescence dosimeter material due to their potential to provide a means of determining cumulative exposure to external gamma radiation arising from accidents or large-scale incidents involving population groups [17]. Moreover, a substantial advantage of this application is that the samples occur in the human body and could thus provide an individual dose estimate for persons who are not equipped with classical personal dosimeters.

In this work, several types of feldspathic ceramics used for the fabrication of the external layer (veneer), which encloses the metal core of the dental crowns, were investigated with respect to their thermoluminescence (TL) and optically stimulated luminescence (OSL) properties for retrospective and accident dosimetry.

2. INSTRUMENTATION

Different types of feldspathic ceramics available on the Romanian dental market (Vita Omega 900 – C1; Vita Titankeramik Opaque – C2, Dentine – C3, Bonder – C4; Carrara Interaction Margin Booster – C5; Duceram SMH High Fusing3 – C6; Duceragold – C7) were analyzed in this work. These materials are most commonly used as veneer porcelains for metal-ceramic restorations.

Veneering ceramics for metal-ceramic restorations, commonly named feldspathic porcelains, are usually leucite-based. Leucite ($KAlSi_2O_6$) is a potassium alumino-silicate that exhibits a tetragonal structure at room temperature and undergoes a displacive phase transformation from tetragonal to cubic at 625°C, accompanied with a volume expansion of 1.2% [20]. Dental porcelain typical oxide composition consists of SiO_2 (~62%), Al_2O_3 (~18%), K_2O (7%), B_2O_3 (~7%), Na_2O (~4%) and other oxides (~2%) [20].

All the investigated samples were used directly from the original packets and measured in form of powder. Each aliquot consisted of a few tens of milligrams of dental ceramic poured in the stainless steel cups placed in the reader's carousel. After measurements, the content of each cup was weighed with a high precision electronic balance for mass normalization, in order to compensate for the variation in the OSL and TL output due to variation in the amount of feldspathic ceramic in each cup. OSL and TL measurements of the unirradiated and irradiated samples (applied doses from 1.5 Gy to 12 Gy) were performed with a Risø TL/OSL Luminescence Reader model TL/OSL-DA-20. The heating rate for TL measurements was 5 °C/s. In order to check for sensitivity changes occurring after repeated cycles of irradiation and readout, two repeat doses (1.5 Gy and 12 Gy) were administered, while the zero-dose (0 Gy) was used to evaluate the recuperation of the signal following multiple irradiation-readout cycles. Optical stimulation was performed using 28 blue LEDs, emitting at 470 nm with a total power of 36 mW/cm^2. The stimulation time was 40 s. The light detection system consists of a bialkali EMI 9235QA photomultiplier tube (PMT), and a UV detection filter (Hoya U-340; 270–370 nm). Irradiations were performed using the built-in $^{90}Sr/^{90}Y$ beta source calibrated using calibration quartz from Risø. Considering that SiO_2 is the main component of dental ceramics, using the same dose rate as for calibration quartz (0.15 Gy/s) is an appropiate approximation. For OSL measurements, the signals used for calculations were integrated over the first 2.4 s of stimulation minus an early background evaluated from 5.4 to 8 s. A similar integration window (of 2 s) was used in a previous study [19] for the investigated fluorapatite veneering glass-ceramic.

3. RESULTS AND DISCUSSION

Typical glow curves of some of the various types of investigated samples unexposed and irradiated with 1.5, 6 and 12 Gy are shown in Fig. 1.

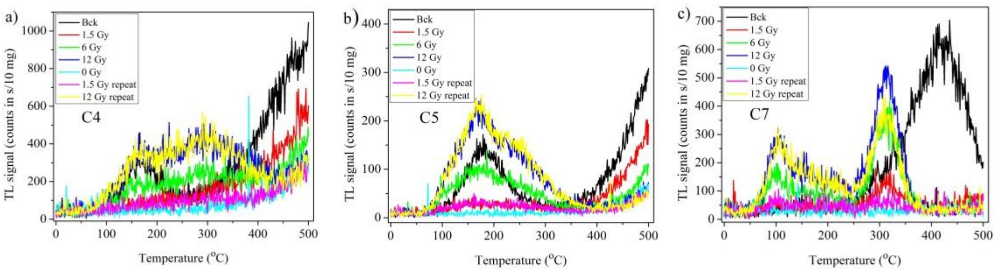

Fig. 1 – Typical TL glow-curves (recorded up to 500°C, heating rate of 5°C/s) measured after irradiation with 1.5 Gy, 6 Gy, 12 Gy. Two repeat doses (1.5 Gy and 12 Gy) were applied in order to check sensitivity variations. The zero-dose was used for verifying signal recuperation. The notation “Bck” means native signal (the background signal from unexposed samples).

All the investigated feldspathic ceramics gave rather weak TL signals, that are generally increasing with the absorbed doses, and complex glow curves characterised by several peaks, features also reported by other authors [17, 18]. At the same time, the glow curve of all samples showed a broad TL peak in the region 100–200°C, which for most of them was followed by a weaker shoulder extending up to approximately 300°C (see as an example C5 and C7 in Fig. 1b and c, respectively). A similar shape of the glow curve was obtained for the feldspathic ceramics investigated by [18], while the dominant TL peak located in the region 100–200°C was reported in [17]. Five of the samples displayed a significant increase of the TL signal occurring in the high temperature region, a feature suggesting the presence of deep traps in the sample matrix (see C4 and C5 in Fig. 1a,b). Only one sample showed the presence of a TL peak at higher temperature, *i.e.* around 300°C, as well as a surprisingly significant peak from the unexposed sample in the region 350–500°C (Fig. 1c). No sensitivity changes or signal recuperation were observed. Regarding native TL signal peak, most samples exhibited a greater or approximately equal peak in comparison to the signals following irradiation in both low and high temperature regions of the glow curve.

The OSL signal of the irradiated samples was recorded for 40 s during continuous wave stimulation using blue light (470 nm with constant power 36 mW/cm^2) and a readout temperature of 125°C. This optical stimulation temperature was used in order to acces traps stable over at least a few hours to several days. Also, no pre-heat treatments were applied. Most of the investigated feldspathic ceramics gave a generally weak OSL signal after irradiation with 1.5 Gy, with values between a minimum of ~100 counts in 0.16 s/Gy for 10 mg of sample in the case of C2 dental ceramic type and a maximum of ~19,000 counts in 0.16 s/Gy for 10 mg corresponding to C7 (Fig. 2). Except for the C7 dental ceramic, the remaining samples were characterised by a slow decay of the OSL signal. A weak intensity of OSL signal was also reported in a previous study [17], while the slow decay rate of the OSL signal was observed by other authors [18]. Also, for most samples the OSL intensity of the native signal was higher than the signal following irradiation with 1.5 Gy (see as an example C3 in Fig. 2b).

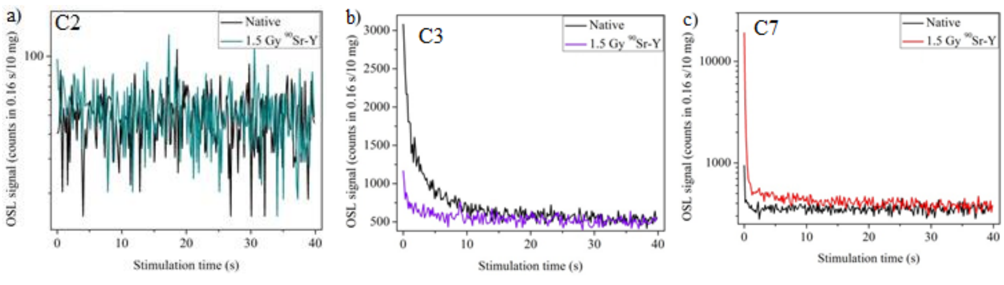

Fig. 2 – Typical OSL decay curves of feldspathic ceramics measured after optical stimulation at 125°C. The OSL signal of unexposed sample (native signal) is compared to that obtained following beta-irradiation with 1.5 Gy.

In order to construct the dose-response relationship, for each sample five doses were applied using the integrated beta source, namely 1.5 Gy, 3 Gy, 6 Gy, 9 Gy, and 12 Gy, followed by the measurement of the signal for a nil dose (0 Gy) and two repeat doses (1.5 Gy and 12 Gy). The growth curve was raised by using the zero-dose signal, as the low values obtained for the nil dose showed insignificant signal recuperation. Most of the samples showed linear OSL dose dependence in the studied dose range (1.5–12 Gy), with $R^2 > 0.99$, except for two types of feldspathic ceramics which were characterised by a cubic behaviour. Figure 3 shows one example for each observed dose dependence. A linear dose response relationship for feldspathic ceramics used in dental restorations was also observed in previous studies [16–19].

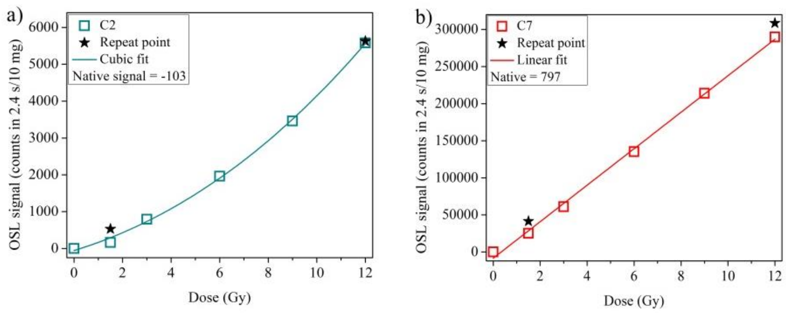

Fig. 3 – Examples of OSL dose response growth curve (a-cubic fit; b-linear fit) of the tested feldspathic ceramics in the dose range of interest (1.5–12 Gy). The star symbols represent the repeat doses delivered in order to check for sensitivity changes.

When applying a single dose protocol corroborated with a readout temperature of 125°C, C7 feldspathic ceramic showed satisfactory features for retrospective dosimetry: intense OSL signal (Fig. 2c), low native signal and a linear dose response relationship, with $R^2 > 0.99$ (Fig. 3c).

As such, we further tested this dental ceramic with regard to its OSL signal repeatability and homogeneity. In order to check the possible presence of sensitivity variations caused by irradiation, we subjected five aliquots to eight repeated irradiation-readout cycles using a constant dose of 3 Gy and no delay time between irradiations and readout. The signal in each measurement cycle was then normalized to the initial response, which was obtained after the first irradiation-readout cycle of each aliquot. The results indicated significant sensitivity variations, showing a constant increase in sensitivity with every irradiation-readout cycle (up to 60% after nine cycles). Afterwards, C7 ceramic homogeneity was tested by irradiating eight aliquots with a constant dose of 6 Gy followed by optical stimulation for 40 s at a readout temperature of 125°C. An average of ~31000 counts in 2.4 s/10 mg was obtained with a relatively high degree of homogeneity (relative standard deviation of 13 %).

Taking into account that this type of feldspathic ceramic is characterised by a relative high degree of homogeneity, but at the same time the OSL signal of each sample irradiated with a dose of 6 Gy increases with each performed measurement, we have concluded that the use of a single aliquot of dental ceramic for measuring the native signal and subsequently constructing a growth curve on the same aliquot would lead to inaccurate results as long as a proper sensitivity change correction protocol (sensus [21]) is not developed and tested.

As this is a very difficult and time consuming task, our following experiments have been designed in a standard multiple aliquot protocol approach corroborated with different stimulation temperatures.

Dose dependence of the OSL signals was studied for doses from 1.5 Gy to 9 Gy and by keeping the investigated samples at different temperatures, namely 25°C, 50°C, 100°C, 125°C, 150°C, and 200°C, during the optical stimulation. For each applied dose, three or five aliquots were used. Consequently, each data point of the growth curves represents the average net signal obtained on three or five aliquots. It was found that C7 feldspathic ceramic exhibited a linear dose-response relationship with high R^2 coefficient of determination values, especially at low stimulation temperatures.

Specific luminescence, *i.e.* luminescence per unit absorbed dose and unit mass, illustrates the sensitivity of the feldspathic ceramic to ionizing radiation, and corresponds to the slope of the dose-signal curves, considering that the curves follow a linear growth for the dose interval of interest. Table 1 summaries the OSL specific luminescence of the investigated feldspathic ceramic as a function of the optical stimulation temperature.

Table 1

OSL specific luminescence of the C7 feldspathic ceramic according to the optical stimulation temperature

Optical stimulation temperature (°C)	OSL specific luminescence (counts in 2.4 s/10 mg)
25	44520 ± 791
50	38731 ± 607
100	30392 ± 729
125	10085 ± 587
150	2782 ± 120
200	6597 ± 448

A significant decrease of the luminescence while increasing the stimulation temperature was observed, indicating that the main and faster component of the OSL signal was due to the recombination process of electrons escaped from shallow traps [18] or to thermal quenching.

When the aliquots native signal was different from zero (with values ranged between ~400 and ~900 counts in 2.4 s/10 mg), the C7 feldspathic ceramic minimum detectable dose (MDD) was estimated as the dose for which the signal is the sum between the mean native signal and three times the standard deviation of the native signal, divided by the OSL specific luminescence of the aliquots (MDD ≃ ≃ $(OSL_{native} + 3\sigma OSL_{native})/c_{specific}$). The MDD was calculated using this formula for most stimulation temperatures: 25°C (room temperature), 50°C, 100°C and 125°C.

Considering that for the remaining stimulation temperatures (150°C and 200°C) the native signal values were negative, the MDD was estimated as the dose for which the signal is three times the standard deviation of the background that corresponds to the last 2.4 seconds of stimulation.

For instantaneous measurements, the obtained values are depicted in Table 2, with the lowest MDD value (9 mGy) corresponding to the 125°C stimulation temperature. However, the actual MDD value would be dependent on the time elapsed from irradiation [19].

Table 2

Minimum detectable dose of C7 feldspathic ceramic as function of the optical stimulation temperature. The values correspond to instantaneous measurements

Optical stimulation temperature (°C)	Minimum detectable dose – MDD (mGy)
25	26
50	50
100	24
125	9
150	307
200	234

These resulting MDD values are lower or similar compared with those found for other common types of dental ceramic [17–19]. The reported MDD for luminescence measurements in previous studies ranged from a few mGy up to a few hundreds of mGy.

Laboratory fading measurements were performed for this feldspathic ceramic by applying a multiple aliquot protocol and various stimulation temperatures (25°C, 125°C and 200°C). The samples were irradiated with a dose of 3 Gy and stored in the darkroom to avoid any light induced signal loss. Optical stimulation at different temperatures was performed both instantaneous and after two delay times following irradiation (12 hours and 48 hours, respectively).

The results indicated a pronounced signal loss during the first 12 hours following irradiation for both 25°C and 125°C optical stimulation temperatures (Table 3). It can be seen that after 12 hours the OSL signal fades almost entirely, remaining only 1.4% of its original value. After this period, the signal loss remained almost constant, with an increase ranged between 0.1% and 2% after 2 days (48 hours) following irradiation.

Table 3

Signal loss determined for C7 dental ceramic after different storage periods. The reported values were normalized to the instantaneous measurements, recorded immediately after irradiation with 3 Gy

Optical stimulation temperatures (°C)	Averaged OSL signal recorded both instantaneous and after varying delay times following irradiation (counts in 2.4 s/10 mg)			Signal loss after different time delays since irradiation (hours)	
	instant	12 hours	48 hours	12	48
25	105303	1439	1377	98.6%	98.7%
125	16724	2047	1735	88%	90%
200	1354	1168	1157	14%	15%

With regard to the highest stimulation temperature (200°C), it can be observed that the fading rate is significantly lowered, the OSL signal being at 85% of its original value after 2 days. Given the fading results, it can be concluded that for dental feldspathic ceramics a high temperature optical stimulation is more advantageous than room temperature stimulation.

Previous studies on dental ceramics also showed that these type of materials exhibit significant fading values in the first few hours post-irradiation followed by an approximately constant fading rate for the next period. Other authors [18] have reported a signal loss of 75% 12 hours post-irradiation when applying a 125°C readout temperature, while after 1 day, in a previous study [19] was observed that the OSL signal measured at room temperature reaches only about 20% of its original value.

4. CONCLUSIONS

Luminescence properties of various feldspathic ceramics used in the manufacture of dental crowns have been investigated with regard to their usefulness as ubiquitous retrospective dosimeters.

The sensitivity to ionizing radiation proved to be strongly depended on the brand of dental ceramic investigated. Most of the samples exhibited weak TL and OSL signals, with high native signals values compared to the 1.5 Gy corresponding signals and a linear, as well as, a cubic dose dependence in the studied dose range (1.5–12 Gy). In comparison with the other brands, C7 feldspathic ceramic exhibited favorable dosimetry characteristics, *i.e.* intense OSL signal, low native signal, a linear dose response relationship, with $R^2 > 0.99$, and a high degree of homogeneity (relative standard deviation of 13%). Moreover, when applying a multiple aliquot protocol and various optical stimulation temperatures, C7 also showed a linear OSL dose dependence, especially at low stimulation temperatures, and a minimum detectable dose ranging from 9 mGy to 300 mGy.

The main limiting factor for the use of C7 dental ceramic in dose reconstruction applications proved to be the fading of the OSL signal to approximately 99% of its original value in the first 12 hours, after applying a 25°C and 125°C optical stimulation. This signal loss is probably due to thermal instability, indicated also by the decrease of the samples luminescence while increasing the stimulation temperature. By contrast, when the OSL signals were recorded after using a 200°C optical stimulation, the samples showed a much slower but constant fading. In the light of these results, a compromise must be made between a slight higher minimum detectable dose (300 mGy), which is not hampering the dosimetric potential of the material, and a much lower fading rate (14%). Therefore, using a multiple aliquot protocol togheter with a 200°C optical stimulation proved to be the suitable measurement procedure for illustrating the C7 dental ceramic usefulness as retrospective dosimeter.

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