



Natural and laboratory OSL growth curve—Verification of the basic assumption of luminescence dating



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HIGHLIGHTS

- Process of OSL growth curve generation in nature and in laboratory was simulated.
- Discrepancies between the natural and the laboratory growth curves are observed.
- Deep disconnected traps play the key role in growth curve inequality.
- Empty deep traps before zeroing of OSL cause the inequality of growth curves.

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ABSTRACT

The basic assumption of luminescence dating is the equality between the growth curve of OSL generated by the natural radiation and the OSL growth curve reconstructed in laboratory conditions. The dose rates that generate the OSL in nature and in laboratory experiments differ by about ten orders of magnitude. Recently some discrepancies between the natural and laboratory growth curves have been observed. It is important to establish their reasons in order to introduce appropriate correction into the OSL dating protocol or to find a test that allows to eliminate the samples which should not be used for dating. For this purpose, both growth curves, natural and laboratory, were reconstructed by means of computer simulations of the processes occurring in the sample during its deposition time in environment as well as those which occur in a laboratory during dating procedure. The simulations were carried out for three models including one shallow trap, two OSL traps, one disconnected deep and one luminescence center. The OSL model for quartz can be more complex than the one used in the presented simulations, but in spite of that the results show effects of growth curves discrepancies similar to those observed in experiments. It is clear that the consistency of growth curves is not a general feature of the OSL processes, but rather a result of an advantageous configuration of trap parameters. The deep disconnected traps play the key role and their complete filling before the zeroing of OSL signal is a necessary condition of the growth curves' consistency.

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1. Introduction

The equality between the curve of the OSL signal growth with radiation dose generated by the natural radiation (natural growth curve) and the OSL growth curve reconstructed in the laboratory conditions (laboratory growth curve) is the basic assumption of luminescence dating and retrospective dosimetry. Both growth curves, however, arise by sample irradiation with extremely different dose rates. In literature there are reports, concerning

mainly the TL of quartz, about the inconsistency of growth curves that are generated with significantly differing dose rates. A short overview of these early works is given by Kvasnička (Kvasnička, 1983). More recently this problem was considered also for the OSL in quartz (Bailey, 2004) and quite recently some doubts have emerged concerning the proper reconstruction of the natural growth curve in laboratory (Timar-Gabor and Wintle, 2013; Timar-Gabor et al., 2012; Timar-Gabor et al., 2015; Chapot et al., 2012). In the cases where there are some discrepancies between both curves, it is important to establish their reasons. This can help to introduce appropriate correction into the OSL dating protocol, which allows to determine the age in problematic cases, or at least to eliminate the sample, which does not give a chance to obtain reliable dating

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outcomes. For a single sample, it is impossible to determine experimentally the natural growth curve, due to the very low natural dose rate and, in consequence, long times of the sample irradiation. Some efforts were made in order to reconstruct the natural growth curve for a sequence of samples originating from sediment profile using the independent age control (Chapot et al., 2012). These measurement revealed that natural- and laboratory-generated growth curves differ in shape. The reason for the growth curves discrepancies, however, is not known. Detailed kinetics studies of the trap filling and the OSL process are needed in order to solve this problem and these can be realized only by means of the computer simulations that involve numerical solution of differential equations, which represent the kinetic model the mentioned processes. In this work, both growth curves, natural and laboratory, were reconstructed and compared for a simplest model that reflects the different kinds of traps existing in quartz. The centers included in this model are: one shallow electron trap (which disrupts the OSL process), two deeper electron traps (active in OSL process), one disconnected deep trap (it does not take part in TL and OSL process) and one luminescence center. The complex set of the recombination centers and hole traps that is known in the case of quartz was omitted, because it is believed that the simplest possible model should be considered at the first stage of the investigations. As it is shown below, such simplified approach brings results that allow to indicate the probable reason of the effects described in the earlier experimental works. The next part of the simulations will be carried out for a widened set of hole traps and luminescence centers.

Two different ways of establishing the kinetics parameters of the model were applied – the one that aimed at the direct reference to known models of OSL in quartz (model 1 and 2) (Bailey, 2001; Adamiec et al., 2006), and the other that was used in order to give more general information about the possible differences between the both growth curves that could concern materials other than quartz (model 3). It should be noted that the problem of nonequivalence of the growth curves generated with different dose rates concerns all the cases in OSL dosimetry when unknown dose is reconstructed by the comparison of the OSL resulting from this dose with the OSL signal generated by a high laboratory dose rate. Hence, the presented results are of wider application than only to the OSL dating.

2. Methods

The simulations have been performed by means of Matlab differential equation solver ode15s, which is the appropriate tool for stiff equations sets. They took into account all the processes occurring in the sample during its deposition time in environment and during laboratory tests. The following steps were included in the simulations for each set of model parameters: 1) natural irradiation with low dose rate 10 a.u. before zeroing the OSL, 2) natural bleaching at 10 °C, 3) natural irradiation at 10 °C, 4) laboratory preheat at 220 °C, 5) natural OSL measurement at 125 °C, 6) laboratory irradiation at 20 °C (high dose rate 3×10^8 a.u., test dose), 7) preheat at 220 °C, 8) laboratory OSL measurement at 125 °C after the test dose, 9) bleaching at 280 °C, 10) laboratory irradiation at 20 °C (high dose rate 3×10^8 a.u., the regeneration dose), 11) preheat at 220 °C, 12) laboratory OSL measurement at 125 °C after the regeneration dose. The kinetic equations solved at each step of simulations have the following form:

$$\frac{dn_i}{dt} = -n_i\sigma_i f - s_i \exp\left(\frac{-E_i}{kT}\right) + A_i(N_i - n_i)n_c; \quad i = 1, \dots, 4 \quad (1)$$

$$\frac{dm}{dt} = A_m(M - m)m_v - \beta mn_c \quad (2)$$

$$\frac{dn_c}{dt} = R + \sum_{i=1}^4 \left[n_i\sigma_i f - A_i(N_i - n_i)n_c + s_i \exp\left(\frac{-E_i}{kT}\right) \right] - \beta mn_c \quad (3)$$

$$\frac{dm_v}{dt} = R - A_m(M - m)m_v \quad (4)$$

where n_i (cm^{-3}) is the concentration of electrons in i -th trap, N_i (cm^{-3}) means the concentration of i -th trap, m (cm^{-3}) is the concentration of holes in recombination centers and M (cm^{-3}) the concentration of these centers, n_c (cm^{-3}) and m_v (cm^{-3}) are respectively the concentrations of free electrons and holes, A_i ($\text{cm}^3 \text{ s}^{-1}$) is the probability of electron trapping in i -th trap, A_m ($\text{cm}^3 \text{ s}^{-1}$) is the probability of hole trapping in the recombination center, R ($\text{cm}^3 \text{ s}^{-1}$) is the intensity of the excitation irradiation producing the pairs of free electron and holes during the trap filling process, β_i ($\text{cm}^3 \text{ s}^{-1}$) is the probability of a free electron recombining with a hole trapped in the luminescence center. The probability of optical excitation of the electron from the i -th trap to the conduction band is equal $\sigma_i f$, where σ_i (cm^2) is the optical cross-section of i -th trap and f ($\text{cm}^{-2} \text{ s}^{-1}$) is the stimulation photon flux. The probability of thermal excitation of the electron from the i -th trap to conduction band is equal $s_i \exp(-E_i/kT)$, where E_i and s_i are the thermal depth and the frequency factor of the i -th trap, respectively. The optical cross-section was calculated for the i -th trap using formula:

$$\sigma_i(h\nu) = \frac{\kappa_i}{\nu\sqrt{\pi}} \int_0^\infty x^2 \exp\left\{ -\kappa_i^2 [x - (h\nu - E_{0i})]^2 \right\} dx \quad (5)$$

$$\kappa_i = \left[2S_i(\hbar\omega_i)^2 \coth\left(\frac{\hbar\omega_i}{2kT}\right) \right]^{-\frac{1}{2}} \quad (6)$$

where $E_{0i} = E_i + S_i\hbar\omega_i/2\pi$ is the optical trap depth, S_i is the Huang-Rhys factor which determines the strength of electron-phonon coupling for the optical transition from the i -th trap to conduction band, $\hbar\omega_i/2\pi$ is the characteristic phonon energy for the i -th trap, $h\nu$ is the stimulation energy and x is a bound variable having the dimension of energy. The calculations of the improper integral are simplified by limiting the integration range to a few electron volts, which does not change the value of $\sigma_i(h\nu)$ noticeably (Chruścińska, 2010). In the case of the model parameters taken from the earlier works presenting OSL models for quartz (model 1 and 2), when only the values of E_i and σ_i were given, the parameters S_i and $\hbar\omega_i/2\pi$ were chosen in such a way that optical cross-sections calculated by means of formulas (5) and (6) were equal to the values given in the literature.

The initial values of parameters used in simulations are listed in Table 1. The concentrations of traps and luminescence centers were changed in order to check their impact on the growth curves. In the simulations for model 3 all the concentrations and retrapping coefficients (A_i) of the traps were equal at the starting point. Next they were changed individually during subsequent tests. In this model the trap depths, frequency factors and parameters determining the electron-phonon coupling are selected, so that the TL peaks position as well the values of the optical cross-sections correspond to the values characteristic for quartz.

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