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On the shape of continuous wave infrared stimulated luminescence signals from feldspars: A case study

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ABSTRACT

The continuous-wave IRSL (CW-IRSL) signals from feldspars are known to decay in a non-exponential manner, and their exact mathematical description is of great importance in dosimetric and dating studies. This paper investigates the possibility of fitting experimental CW-IRSL curves from a variety of feldspar samples, by using an analytical equation derived within the framework of a new model based on localized electronic recombinations of donor–acceptor pairs. 24 different types of feldspars were studied and their CW-IRSL signals are analyzed in order to establish the range and precision of numerical values for the fitting parameters in the analytical equation. The study finds systematic trends in the fitting parameters, and possible systematic differences between K and Na rich extracts from the same feldspar samples. Furthermore, results are compared with natural samples, freshly irradiated samples, and samples which had undergone anomalous fading. The results of this analysis establish broad numerical ranges for the fitting parameters in the model. Specifically the possible range of the dimensionless density ρ' was found to be $\rho' \sim 0.002–0.01$. These experimentally established ranges of ρ' will help to guide future modeling work on luminescence processes in feldspars. Small statistical differences were found between K-rich and Na-rich fractions of the same sample. However, the experimental data shows that the parameters depend on the irradiation dose, but do not depend on the time elapsed after the end of the irradiation process. All samples exhibited the power law of decay, with the range of the power law coefficient $\beta = 0.6–1.1$.

1. Introduction

Over the past 20 years there has been considerable experimental and modeling work trying to understand the nature and properties of luminescence signals from feldspars, especially in connection with the associated phenomenon of “anomalous fading” based on quantum mechanical tunneling (see for example [1–3]). The continuous-wave IRSL signals (CW-IRSL, also known as “infrared shine down” curves) from these materials are known to decay in a non-exponential manner and their origin as well as their exact mathematical description is an open research question. Mathematical and physical characterization of the shape of these CW-IRSL signals in feldspars is of great importance in dosimetric and dating studies (Refs. [4–32]).

There is an abundance of experimental data as well as significant modeling work in the literature which suggests that the dominant process in anomalous fading in feldspars is tunneling from the ground state or via the excited state, or from both. Poolton et al. [7–8,17] explained IRSL in feldspars using a donor–acceptor model, in which electron tunneling occurs from the excited state and the band tail states of the IRSL trap at about 1.4 eV. Thomsen et al. [22,23] suggested that in this model, the beginning of the IRSL decay curve originates with the luminescence emitted from close donor–acceptor pairs, while the end of the IRSL curve most likely represents the tunneling of distant pairs. The recent experimental work by Poolton et al., [17], Jain and Ankjærgaard [18], Ankjærgaard et al. [19], Andersen et al. [31] and Kars et al. [32] provided valuable information on the origin of these CW-IRSL signals and support the presence of several competing mechanisms during the luminescence process. These mechanisms involve the tunneling processes in localized recombinations taking place from the ground and excited state of the trap, as well as charge migration through the conduction band-tail states.

There have also been significant developments in the modeling aspects of the luminescence mechanism for CW-IRSL signals, including the experimentally observed empirical power law of decay in feldspars ([14–16]). Kars et al. [11] and Larsen et al. [12] presented new types of models involving localized tunneling transitions, while Pagonis et al.

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[20] presented a new empirical kinetic model, in an attempt to describe tunneling via the excited state in feldspars. In an important development in this research area, Jain et al. [21] presented a new general kinetic model which quantifies localized electronic recombination of donor–acceptor pairs in luminescent materials. Recombination is assumed to take place via the excited state of the donor, and to take place between nearest-neighbors within a random distribution of centers. Two versions of the model were presented: an exact model that evolves in both space and time, and an approximate semi-analytical model that evolves only in time. These authors found good agreement between the two models, and simulated successfully both thermally stimulated luminescence (TL) and optically stimulated luminescence (OSL). The model also demonstrated the power law behavior for OSL signals simulated within the model.

Pagonis et al. [33] examined the full model by Jain et al. [21], and obtained analytical expressions for the distribution of remaining donors at any time t during the following experimental situations: TL, OSL, linearly modulated OSL and infrared stimulated luminescence (LM-OSL, LM-IRSL) and isothermal TL (ITL). These authors gave examples for the derived distributions of donors in each experimental case, and similarities and differences between the different experimental modes of stimulation were pointed out. They also demonstrate how LM-IRSL signals in feldspars can be analyzed using the model, and what physical information can be extracted from such experimental data. Their analytical equations were tested by fitting successfully a series of experimental LM-IRSL data for Na- and K-feldspar samples available in the literature.

Kitis and Pagonis [34] showed that the system of simultaneous differential equations developed by Jain et al. [21] can be approximated to a very good precision by a single differential equation describing stimulated luminescence emission in this system. These authors were able to obtain analytical solutions of this single differential equation for several possible modes of stimulation, namely TL, OSL, LM-OSL and ITL. They also derived the exact analytical form for the power law behavior in this system, and demonstrated how typical experimental TL glow curves and CW-IRSL signals can be analyzed using the derived analytical equations, and what physical information can be extracted from such experimental data.

The goals of the present paper are as follows.

(a) To investigate the possibility of fitting experimental CW-IRSL curves from a variety of feldspar samples by using the analytical equation derived by Kitis and Pagonis [34] within the framework of the model by Jain et al. [21].
(b) To investigate the range of numerical values, as well as the precision of the kinetic parameters derived from fitting experimental data to the analytical equation derived by Kitis and Pagonis [34]. Special attention is paid in looking for systematic trends in the fitting parameters, and for possible systematic differences between K and Na rich extracts from the same feldspar samples.
(c) To compare the fitting parameters obtained from CW-IRSL curves measured in 3 types of samples: natural samples, freshly irradiated samples, and samples which have undergone anomalous fading. The specific goal here is to search for possible effects of laboratory irradiation and anomalous fading on the shape of the CW-IRSL signals.
(d) To examine the power law decay of these signals from different types of feldspars.

2. Analytical solution for CW-IRSL signals within the model of Jain et al. [21]

In this section we summarize briefly the main physical assumptions and equations used in the model of Jain et al. [21], and discuss the analytical equation used in this paper to fit experimental CW-IRSL curves. The main physical assumption in the model of Jain et al. [21] is the presence of a random distribution of hole traps in the luminescent volume, and an associated range of random nearest-neighbor recombination probabilities. Within the model, stimulated recombination takes place only via the excited state of the electron trap, by either optical or thermal stimulation. The concentration of holes is assumed to be much larger than the population of electrons in traps so that there is no redistribution of electron–hole distances during the experiment, and an electron can tunnel only to its nearest hole. In the exact form of the model, one writes a system of differential equations describing the traffic of electrons between the ground state, the excited state and the recombination center. These coupled differential equations contain two variables, namely the distance r' between donor–acceptor pairs and the time t.

Jain et al. [21] also developed an approximate semi-analytical model to describe the behavior of the system. This second version of the model evolves only in time, and the approximation used is based on introducing a critical tunneling lifetime τ_c. The equations in the approximate semi-analytical model version of the model and for the case of CW-IRSL experiments are [21]as follows:

$$\frac{dn_g}{dt} = -An_g + Bn_e, \tag{1}$$

$$\frac{dn_e}{dt} = An_g - Bn_e - \frac{3n_e\rho'^{1/3}}{\tau_c}\left(\ln\frac{n_o}{n}\right)^{2/3} z, \tag{2}$$

$$L(t) = -\frac{dm}{dt} = \frac{3n_e\rho'^{1/3}}{\tau_c}\left(\ln\frac{n_o}{n}\right)^{2/3} z, \tag{3}$$

$$\tau_c = s^{-1}\exp\left[\left(\frac{1}{\rho'}\ln\frac{n_o}{n}\right)^{1/3}\right]. \tag{4}$$

The following parameters and symbols are used in these expressions: n_g and n_e are the instantaneous concentrations of electrons in the ground state and in the excited state correspondingly. m is the instantaneous concentration of acceptors (holes), n is the instantaneous concentration of all the donors, and N represents the instantaneous concentration of electrons in thermally disconnected states, such that $m = n + N = (n_g + n_e) + N$. The parameter A represents the excitation rate from the ground to the excited state, and is equal to $A = \sigma(\lambda)I$ for the case of optical excitation. Here λ is the optical stimulation wavelength, $\sigma(\lambda)$ is the optical absorption cross-section and I is the light intensity ($cm^{-2}\,s^{-1}$). Additional parameters are the dimensionless number density of acceptors ρ', the critical tunneling lifetime τ_c, the thermal excitation frequency factor s, and $z = 1.8$ – a dimensionless constant introduced in the model.

B (s^{-1}) is the relaxation rate from the excited into the ground state, and $L(t)$ is the instantaneous tunneling luminescence from recombination via the excited state. Under the detailed balance principle one also has $B = s$. Perhaps the most important physical parameter in the model is the dimensionless number density of acceptors ρ' and the critical tunneling lifetime τ_c which depends on the instantaneous concentration of donors n as shown in Eq. (4). Jain et al. [21] simulated successfully both TL and optically stimulated luminescence (OSL) processes in their model, and also demonstrated the power law behavior for simulated OSL signals within the model. However, their approximate semi-analytical model was found to disagree with the exact solution of the model in the case of low values of the optical excitation probabilities $A = 1$–$10\ s^{-1}$ (Ref. [21, Fig. 7a and 7b]).

Kitis and Pagonis [34] showed that under certain simplifying physical assumptions, the system of Eqs. (1)–(4) can be replaced