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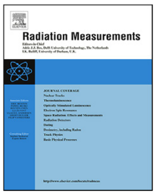

Radiation-induced growth and isothermal decay of infrared-stimulated luminescence from feldspar

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HIGHLIGHTS

- We review models of dose response and isothermal decay in feldspar IRSL.
- We promote a uniform visualisation of these phenomena on a log(time) scale.
- We examine a general-order kinetics model successfully describing both phenomena.
- We benchmark all models against a previously published MET-pIRIR dataset.

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ABSTRACT

Optically stimulated luminescence (OSL) ages can determine a wide range of geological events or processes, such as the timing of sediment deposition, the exposure duration of a rock surface, or the cooling rate of bedrock. The accuracy of OSL dating critically depends on our capability to describe the growth and decay of laboratory-regenerated luminescence signals. Here we review a selection of common models describing the response of infrared stimulated luminescence (IRSL) of feldspar to constant radiation and temperature as administered in the laboratory. We use this opportunity to introduce a general-order kinetic model that successfully captures the behaviour of different materials and experimental conditions with a minimum of model parameters, and thus appears suitable for future application and validation in natural environments. Finally, we evaluate all the presented models by their ability to accurately describe a recently published feldspar multi-elevated temperature post-IR IRSL (MET-pIRIR) dataset, and highlight each model's strengths and shortfalls.

1. Introduction

Optically stimulated luminescence (OSL) dating of feldspar, commonly utilising stimulation with infrared (IR) light and hence termed IRSL, is a group of methods enabling the determination of depositional ages of middle to late Quaternary sediments (Hütt et al., 1988; Buylaert et al., 2012; Li et al., 2014). More recently, the geological applications of feldspar IRSL have been extended to surface exposure dating (Sohbati et al., 2011) and low-temperature thermochronology (Guralnik et al., in review). In addition to the chemical or physical characterisation of a sample's natural radioactivity, the conversion of its natural luminescence into a radiometric age involves two laboratory experiments, in which the luminescence is monitored as a function of the exposure time t [s] to (i) a source of constant radioactivity $\dot{D}$ [Gy s^{-1}], and (ii) a source of a

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constant temperature T [K]. The former experiment determines how fast does the luminescence signal grow under an artificial radiation source, and the latter (often skipped in routine sediment dating) quantifies the thermal stability of the dosimetric electron trap.

Although the observable rates of luminescence growth and decay in the laboratory are typically faster by a factor of ~10^{10} than in nature, geological dating must assume that the kinetic parameters describing laboratory behaviour are fundamental physical characteristics of the material, that can be extrapolated over much longer timescales and slower rates. Thus, the selection of a model for describing laboratory behaviour is more than critical for the correct and meaningful conversion of the natural luminescence intensities into equivalent ages. Even if a model produces an excellent fit to laboratory data, this cannot necessarily guarantee its successful extrapolation to geological timescales; at the same time, a model which does not fit laboratory data is even harder to evaluate, since it may further propagate this failure unpredictably, potentially yielding correct ages even though the model is inadequate. In this paper, we take a fresh look at the conventional 'status quo' models currently used to describe dose response and thermal sensitivity of feldspar IRSL. We further examine an interesting heuristic approach (the General-Order Kinetic model), and use a representative dataset to graphically illustrate the key differences between the models, and to quantify their relative successes and shortfalls.

2. Data and methods

2.1. Feldspar MET-pIRIR dataset

The various models discussed in this paper were tested against data that was obtained using the multi-elevated temperature post-IR IRSL protocol (MET-pIRIR; Li and Li, 2011). This protocol retrieves five different IRSL signals measured at incrementally rising stimulation temperatures (50, 100, 150, 200 and 250 °C), and typically exhibiting different thermal stabilities. The specific dataset used in our study, is taken from the work of Li and Li (2012, 2013), and is provided as a digital appendix for any future re-evaluation (see Supplementary material). The data for each of the five post-IR signals (abbreviated MET-pIRIR$_x$, where x is the stimulation temperature) consists of a radiation-induced luminescence growth experiment (a single time-series, observed at a room temperature of ~15 °C), and an isothermal luminescence decay experiment (four individual time-series, measured at temperatures of 300, 320, 340 and 360 °C, and fitted simultaneously).

2.2. Fitting and smoothing procedures

Nonlinear least-square fitting and estimation of errors was performed using the *lsqnonlin* and *nlparci* functions in Matlab. Trends in the fitting residuals (Fig. 1) and in the best-fit parameters (Fig. 3) were visualised using the locally weighted regression and smoothing (LOWESS) method of Cleveland (1979).

2.3. Data visualisation

An implicit tradition in modern OSL literature (e.g. Murray and Wintle, 2000) stipulates the presentation of radiation-induced luminescence growth in form of a 'dose-response' curve, in which the luminescence light sum $L(t)$ varies as a function of the 'absorbed dose' $D = \dot{D}t$ (e.g. Fig. 1a–d). Conversely, isothermal luminescence decay experiments are typically visualised as $log(L(t)/L_0)$ against time t only (e.g. Murray and Wintle, 1999). In the present paper we use a slightly modified visualisation scheme (after Levy, 1961, 1991; Li and Li, 2013), in which the luminescence intensity $L(t)$ is always plotted against $log(t)$ regardless of whether luminescence growth or decay are being explored. The specific benefits of this scheme are:

(i) *Separation of data from interpretation*. When luminescence $L(t)$ is plotted against the absorbed dose $D = \dot{D}t$, the x-axis unnecessary entangles a primary observation (irradiation time t) with a derived parameter (the dose rate $\dot{D}$), the latter incorporating multiple internal and external uncertainties (Bos et al., 2006; Guérin et al., 2011; Kadereit and Kreutzer, 2013; Boehnke and Harrison, 2014). Thus, a plot of $L(t)$ vs. D technically becomes erroneous with every systematic revision of dose rate conversion factors, while a plot of $L(t)$ vs. t will not only remain valid, but also be easier to re-analyse in the future. Furthermore, it is well-known that in materials suffering from athermal losses, delivery of the same dose at different irradiation rates leads to differential luminescence responses (e.g. Kars et al., 2008). Thus, showing luminescence response against an amalgamated variable which is the product of both time and dose rate $D = \dot{D}t$ leads to misapprehension of the dependence of luminescence build-up on laboratory dose rate (see Levy (1961), (1991)).

(ii) *Visual informativeness*: The processes of luminescence growth and decay are both governed by a fundamental rate term [s^{-1}], which drives each corresponding process towards a secular steady-state. Derivation of reliable kinetic parameters typically relies on data which is uniformly spaced across 3–4 orders of magnitude of time (e.g. Kars et al., 2008; Murray et al., 2009; Timar-Gabor et al., 2012). Thus, the use of a linear time axis may unfavourably compress information from a particular timescale, and lead to a visual misapprehension of the fit quality, or of the lack of experimental points to prove or disprove a certain model (compare Fig. 1a–d with Fig. 1e–h, showing exactly the same data $L(t)$ but as a function of $D = \dot{D}t$ and $log(t)$, respectively). The above problems are less likely to occur on a logarithmic time axis $log(t)$, which not only grants easy comparison between similar processes occurring on different timescales, but also highlights regions where data is missing to properly constrain the model fitting

(iii) *Uniformity for internal comparison*: Visualisation of luminescence growth and decay as a function of $log(t)$ allows a straightforward side-by-side comparison of the kinetic responses of the material to cumulative irradiation and heat, and in both cases facilitates the detection and quantification of systematic departure from first-order kinetics (see Section 3.3 and Fig. 2). Although the proposed visualisation might be slightly difficult to compare to former studies (utilising the traditional plotting approach), we believe that this is a minor inconvenience outweighed by the benefits of internal intercomparison, and of an enhanced apprehension of model quality.

3. Models and results

3.1. First-order (exponential) kinetics (1EXP)

The growth of the IRSL light sum $L(t)$ [a.u.] in a feldspar exposed to a radioactive source may be described by a saturating exponential function:

$$L(t)/L_{\max} = 1 - \exp(-\dot{D}t/D_0) \quad (1)$$

(e.g. Balescu et al., 1997; Li and Li, 2012) where L_{max} [a.u.] is the maximum luminescence light sum, $\dot{D}$ [Gy s^{-1}] the constant dose