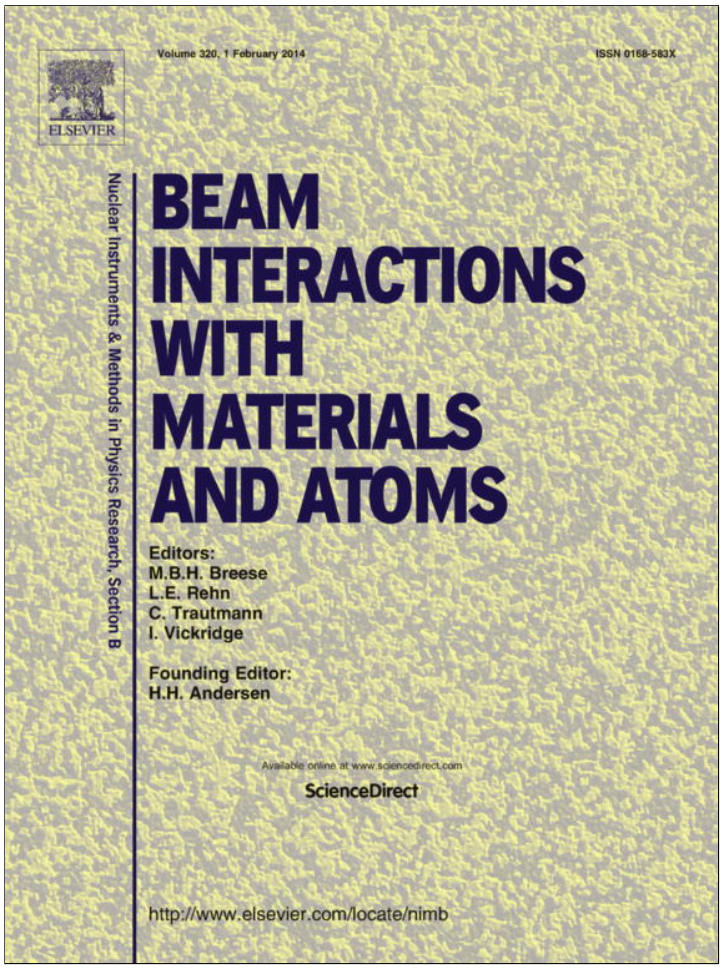

Nuclear Instruments and Methods in Physics Research B 320 (2014) 57–63

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

# Prompt isothermal decay of thermoluminescence in an apatite exhibiting strong anomalous fading

I.K. Sfampa [a], G.S. Polymeris [b,d], N.C. Tsirliganis [d], V. Pagonis [c], G. Kitis [a,*]

[a] Aristotle University of Thessaloniki, Nuclear Physics Laboratory, 54124 Thessaloniki, Greece
[b] Aristotle University of Thessaloniki, Solid State Physics Section, 54124 Thessaloniki, Greece
[c] McDaniel College, Physics Department, Westminster, MD 21157, USA
[d] Laboratory of Radiation Applications and Archaeological Dating, Department of Archaeometry and Physicochemical Measurements, 'Athena' R. & I. Center, Kimmeria University Campus, GR67100 Xanthi, Greece

ARTICLE INFO

*Article history:*
Received 1 July 2013
Received in revised form 6 November 2013
Available online 28 December 2013

*Keywords:*
Thermoluminescence
Anomalous fading
Apatite
Tunneling luminescence
Luminescence dating
Isothermal TL

ABSTRACT

Anomalous fading (AF) is one of the most serious drawbacks in thermoluminescence (TL) and optically stimulated luminescence (OSL) dating. In the present work the isothermal decay of TL signals from Durango apatite is studied for temperatures located on the rising part of the main TL peak. This material is known to exhibit strong AF phenomena, and its isothermal TL decay properties have not been studied previously. The experimental results show that the characteristic decay time of the isothermal signal does not depend of the temperature, and that this signal does not exhibit the strong temperature dependence expected from conventional TL kinetic theories. This is further direct experimental evidence for the possible presence of tunneling phenomena in this material. The isothermal decay curves are analyzed and discussed within the framework of conventional theories of TL, as well as within the context of a recently developed tunneling kinetic model for random distributions of electron-hole pairs in luminescent materials.



## 1. Introduction

In some inorganic materials a rapid fading of the luminescence signals is observed experimentally within short times after irradiation, instead of within the long lifetimes predicted by standard kinetic models. This rapid fading process has been termed anomalous fading (AF), and is one of the most serious problems in TL and OSL dating [1,2].

Explanations of the AF effect have been based on various proposed models, such as the tunneling model [3–5], the localized transition model [6,7] and a model based on competition with radiationless transitions [8]. Currently, the most accepted explanations of AF are based on quantum mechanical tunneling from the ground state or from the excited state of the trap [9–13].

For the investigation and understanding of the anomalous fading effect it is desirable to find experimental conditions and sample preconditioning which would strongly influence the AF phenomena. Durango apatite is a natural material which is known to exhibit strong anomalous fading effects [14], with AF in this material exhibiting a remarkable resistance to a variety of experimental conditions and sample preconditioning. Kitis et al. [15] studied the effect of varying the heating rate during a TL measurement on the AF properties of this crystal [16,17], while Tsirliganis et al. [18] investigated the dependence of the AF of TL/OSL signals in this material on the occupancy of the recombination sites. While the influence of AF on the TL and OSL signals has been studied extensively for a variety of materials, the effect of AF on the isothermal TL signals has not been studied previously in a quantitative manner.

In a review article, Visocekas [19] discussed the nature and experimental importance of tunneling afterglow signals observed in a variety of materials. Baril [20] and Baril and Huntley [21] carried out detailed isothermal experiments in feldspars and showed that at prolonge times the isothermal signals follow a power law, with a power law coefficient very close to 1.

The general aim of the present work is to investigate the isothermal decay signals from Durango apatite, and the possible influence of AF on these isothermal signals. The specific goals of this work are:

- To analyze the isothermal luminescence signals using several standard methods of analysis.
- To study the effect of AF on isothermal signals. This was achieved by comparing isothermal signals measured immediately after irradiation, with signals measured 24 h after irradiation.
- To discuss the experimental results within the framework of a recently proposed model which is based on localized transitions and tunneling.

* Corresponding author. Tel.: +30 2310998175.
*E-mail address:* gkitis@auth.gr (G. Kitis).

0168-583X/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.nimb.2013.12.003

## 2. Experimental procedure

### 2.1. Sample details and preparation

The sample for these experiments was a natural crystal of Durango apatite with dimensions of $8 \times 4 \times 3$ mm. The single piece crystal was crushed gently with an agate mortar and grains of dimension 80–140 μm were obtained after sieving. The grains were annealed at 900 °C for 1 h, followed by rapid cooling to room temperature. This annealing treatment is necessary in order to empty all traps, which have been filled by the natural irradiation of the material. Previous work has shown that this annealing process does not influence the anomalous fading effect in Durango apatite [14].

Aliquots (sub-samples) with the same mass of 5 mg were attached to stainless steel disks. Each data point reported in this paper was the average of two measurements carried out on two different aliquots/disks.

### 2.2. Apparatus and measurement conditions

TL measurements were carried out using a Risø TL/OSL reader (model TL/OSL–DA–15), equipped with a $^{90}Sr/^{90}Y$ beta particle source, delivering a nominal dose rate of 0.075 Gy/s. A 9635QA photomultiplier tube with a combination of Pilkington HA-3 heat absorbing and Corning 7–59 (320–440 nm) blue filter were used for light detection. All measurements were performed in a nitrogen atmosphere with a low constant heating rate of 2 °C/s, in order to avoid significant temperature lag, and the samples were heated up to the maximum temperature of 500 °C. There are two experimental methods to study the decay of TL under stable temperature. (a) The Residual isothermal decay method (RID) in which the irradiated sample is post irradiation annealed in a furnace and then the residual TL glow-curve is measured. (b) The prompt isothermal decay (PID) method in which TL is measured directly while the sample is held to a stable temperature in the TL reader. The PID method is used in the present work.

### 2.3. Experimental protocols

The experimental procedure for the PID study of TL was performed according to the following protocol.

- Step 1: The previously annealed aliquot is irradiated with a test dose TD = 15 Gy, in order to populate the traps and centers.
- Step 2: TL measurement up to a temperature $T$ at 2 °C/s. At this temperature, called $T_{dec}$ the sample is left to decay thermally for 1000 s.
- Step 3: After the end of the decay period the sample is cooled down to room temperature.
- Step 4: Repeat steps 1–3 for a new aliquot and for a new decay temperature $T_{dec}$.

The prompt isothermal TL decay temperatures $T_{dec}$ used in step 2 are shown in Fig. 1. The glow curve yields a small peak around 140 °C and the main TL peak is located at 310 °C. The $T_{dec}$ values range from 160 °C up to 250 °C in steps of 10 °C. The starting $T_{dec}$ of 160 °C was chosen to erase sufficiently the low temperature TL, and the highest $T_{dec}$ of 250 °C is just below the temperature corresponding to half of the maximum TL intensity. Higher $T_{dec}$ values were avoided for the following reasons. (a) The percentage of trapped electrons thermally released during the TL readout up to $T_{dec}$ must be kept in a relatively low level, in order for the isothermal decay to represent accurately the decay of the whole population of trapped electrons. (b) The thermal decay constant is expected to depend very strongly on temperature $T_{dec}$, so that at temperatures close to, or higher than, the peak maximum temperature the isothermal decay will become extremely short lived and difficult to measure.

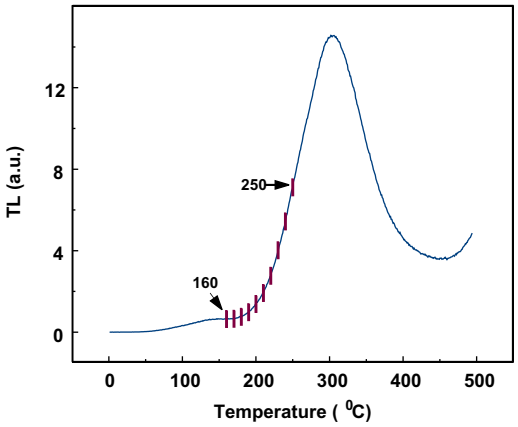


**Fig. 1.** Glow-curve of Durago apatite. The lines indicates the position on the glow-curve of the PID temperature used, in steps of 10 °C.

The above protocol was run twice. In the first run the TL measurement in step 2 was performed immediately after the end of irradiation in step 1. This means that the time left for anomalous fading is practically zero, so this case will be referred as "zero storage" case. In the second run the sample after the irradiation in step 1 was stored at room temperature for 24 h. During the 24 h storage period a substantial amount of anomalous fading has taken place, reducing the TL intensity by about 50% [16,17]. Longer storage times cause a substantial TL signal loss, making a PID experiment statistically poor.

The application of the experimental protocol produces two sets of prompt isothermal decay curves. In the "zero storage" set the isothermal decay curve will represent the thermal decay effect without a serious loss of TL signal due to anomalous fading. The second 24 h storage set of measurements will represent a substantial loss of TL signal due to the anomalous fading, and will be referred to as the "24 h storage" measurements.

## 3. Results

### 3.1. Analysis based on exponential functions

The decay curves obtained from the PID experiment for "zero storage" time are shown in Fig. 2(a). The decay curves for the "24 h storage" samples were very similar and are not shown here. Due to the very different intensity at each $T_{dec}$, the curves are normalized over the initial intensity at time $t = 0$. The results of Fig. 2(a) are unexpected, in the sense that they contradict well–established TL theories based on delocalized transitions through the conduction band. As is well known from TL kinetic theory [22,23], the decay constant $\lambda$ (T) is expected to depend very strongly on $T_{dec}$. However, the PID curves in Fig. 2(a) do not follow this expected behavior.

An example of the expected typical behavior of experimental PID curves in dosimetric materials is given in Fig. 2(b) for the case of $Al_2O_3$:C. As an additional example based on simulation, thirteen (13) pairs of $(E, s)$ values were found which would give the same peak maximum temperature $T_m$ at 300 °C. Using these pairs of parameters, the PID curves were simulated at the temperatures of Fig. 1. A typical result of the simulation is shown in Fig. 2(c), showing clearly a different behavior than the experimental data in Fig. 2(a).

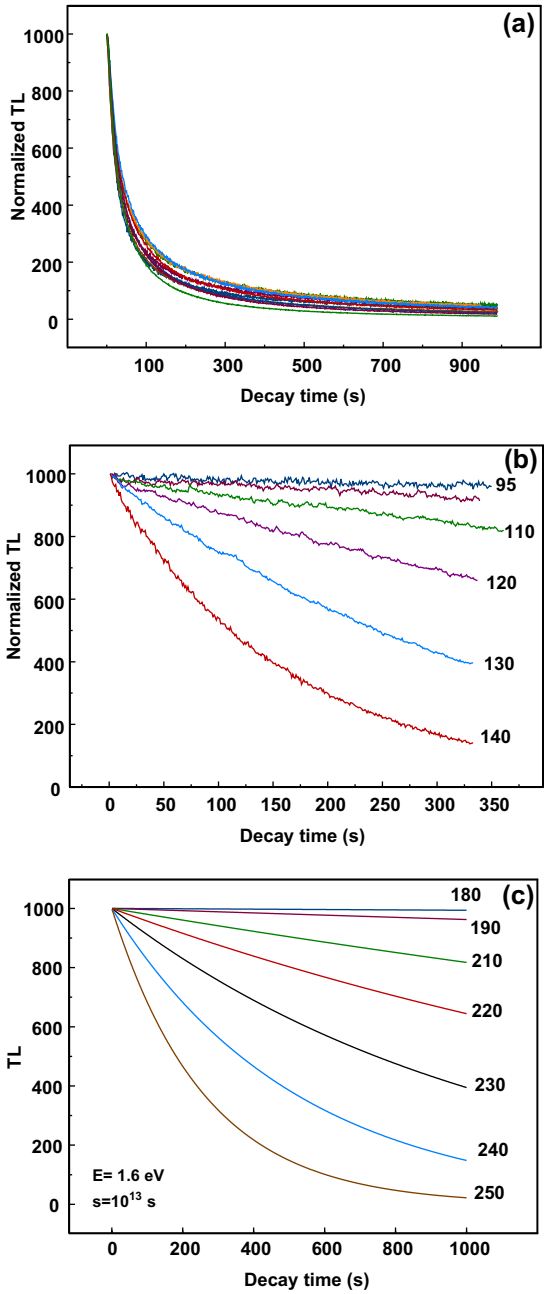


**Fig. 2.** (a) Normalized PID curves received at the decay temperatures shown in Fig. 1. (b) A PID example for $Al_2O_3$:C and (c) a simulation example for a peak having the same peak maximum as the Durango apatite, for the PID temperatures shown in Fig. 1.

Although it is clear that the PID curves in Fig. 2(a) do not follow the expected behavior of TL signals from delocalized models, it is important to analyze the experimental results using standard methods of analysis, in order to show the discrepancies between theory and experiment in a quantitative manner.

### 3.1.1. Analysis based on first order kinetics

The most common method of analyzing PID curves is by using a linear combination of exponentials representing components following first order kinetics. The first order kinetics fit of the PID curves of Fig. 2(a) was achieved using two exponential components termed $C_1$ and $C_2$, plus a stable component which was left to vary during the fitting procedure. The goodness of fit was tested by the figure of merit (FOM) [24]. All fits of the experimental decay curves for both "zero storage" and "24 h storage" gave excellent results with FOM values ranging from 3% at $T_{dec}$ 160 °C to 1% at $T_{dec}$ 250 °C.

The resulting values of $\tau$ from this fitting procedure are given in Table 1. The values of $\tau$ corresponding to the components $C_1$ and $C_2$ for all cases were $26.8 \pm 4$ s and $189 \pm 20$ s, correspondingly. The relative contributions of the components $C_1$ and $C_2$ and of the stable component were 54%, 20% and 25%, correspondingly, with a slight temperature dependence for the constant component.

The conclusion of this analysis based on exponentials is that the PID curves can be described by two exponentials with lifetimes $\tau$ which are independent of the temperature used during the isothermal experiment. This is a result in clear contradiction of standard TL models based on delocalized transitions, in which one expects a strong dependence of the lifetime on the temperature of the isothermal TL experiment.

### 3.1.2. Analysis based on general order kinetics

Using another standard method of analysis, the PID curves were analyzed using the general order kinetics expression for isothermal TL [25],

$$I(t) = I_0 \left[1 + (b-1)\,\frac{t}{\tau}\right]^{-\frac{b}{b-1}}, \quad b \neq 1, \tag{1}$$

where $I(t)$ is the intensity of the luminescence signal as a function of time, $\tau = 1/\lambda$ (s) is the lifetime and $b$ is the order of kinetics. For the case of general order kinetics the kinetic order $b$ was left to vary freely up to the value of $b = 2.05$. The fits obtained for the experimental decay curves of both "zero storage" and "24 h storage" samples were excellent with FOM values ranging from 3% at $T_{dec}$ 160 °C to 1% at $T_{dec}$ 250 °C. The value of the kinetic order was always found to be around $b \sim 2$. The resulting values of $\tau$ are given in Table 1. Specifically, the values of $\tau$ corresponding to the first general order component were $\tau_1 = 65 \pm 6$ s and $\tau_1 = 76 \pm 7.6$ s for "zero storage" and "24 h storage" samples correspondingly. According to these results, there is a small difference in the $\tau_1$ values of the two samples, however this effect seems to be within the statistical accuracy of the experiments.

The values of $\tau$ for the second general order component show a dependence on temperature, as shown in Fig. 3. The results of Fig. 3 indicate the possible presence of a thermally activated process.

The analysis also gave a relative stable percentage of each component as a function of $T_{dec}$. Specifically the percentages for "zero storage" were $52.5 \pm 8$ and $47 \pm 5$ for the two general order components correspondingly, whereas for "24 h storage" samples they were $40.0 \pm 8$ and $60 \pm 5$, respectively.

The integrated signal of the PID curve shows a very strong increase as a function of $T_{dec}$ as shown in Fig. 4.

Based on the above analysis, one can conclude that the general order kinetics analysis can neither explain the observed experimental PID curves of Fig. 2(a). Other possibilities are examined in the next two subsections.

## 3.2. Analysis based on tunneling plus exponential components

The anomalous fading exhibited by Durango apatite is a strong indication that a tunneling mechanism must be considered as a possible explanation of the isothermal results. This section focuses on the recent work by Jain el al [26]. 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-neighbor recombination probabilities. Stimulated recombination takes place only via the excited state of the electron trap, by either optical or thermal

**Table 1**
Results of analysis of PID curves using several different methods of analysis. The $\tau$ values are in seconds.

| Exponential analysis | | | |
|---|---|---|---|
| *First order kinetics* | | | |
| Storage time | $\tau_1$ | $\tau_2$ | Component 3 |
| "zero" and 24 h | $26.8 \pm 4$ | $189 \pm 20$ | Stable |
| *General order kinetics* | | | |
| Storage time | $\tau_1$ | $\tau_2$ (Temperature dependent) | |
| "Zero" | $65 \pm 6$ | Fig. 3 | |
| 24 h | $76 \pm 7.6$ | Fig. 3 | |
| Tunneling plus exponential analysis | | | |
| *Tunneling component* | | | |
| Storage time | $\tau$ | $\rho'$ | % |
| "zero" | $0.924 \pm 0.012$ | $0.0019 \pm 0.0004$ | $74.5 \pm 8.1$ |
| 24 h | $0.995 \pm 0.02$ | $0.002 \pm 0.0004$ | $59.3 \pm 9.8$ |
| *Exponential component (temperature dependent)* | | | |
| Storage time | $E(eV)$ | Figure | % |
| "zero" | $0.776 \pm 0.05$ | Fig. 6 | $20.8 \pm 3.0$ |
| 24 h | $0.704 \pm 0.04$ | Fig. 6 | $37.1 \pm 8.0$ |
| Tunneling analysis | | | |
| Storage time | $\tau$ | $\rho'$ (temperature dependent) | % |
| "zero" | $1.14 \pm 0.14$ | Fig. 8 | over 95% |
| 24 h | $1.25 \pm 0.1$ | Fig. 8 | over 90% |

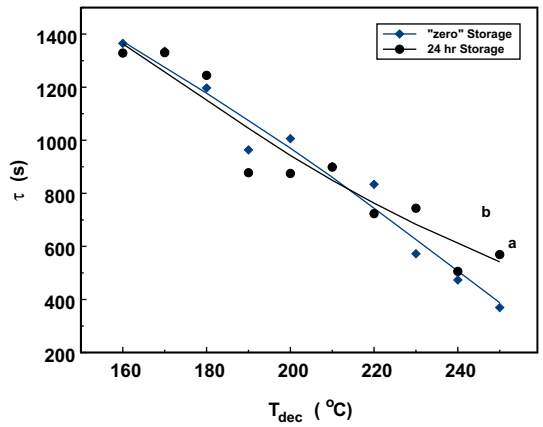


**Fig. 3.** Lifetime $\tau$ of the second exponential component obtained from the analysis described in Section 3.1.2.

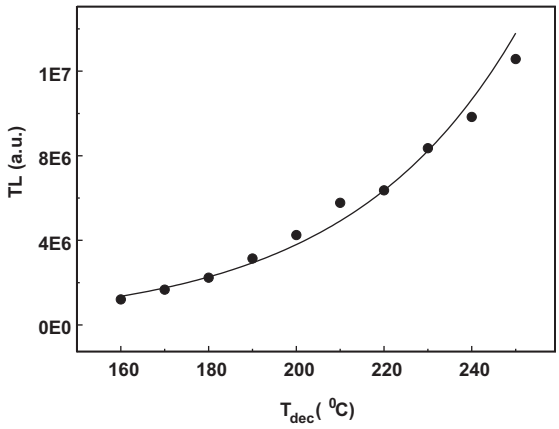


**Fig. 4.** Behavior of the TL integrated signal under each PID curve as a function of $T_{dec}$.

stimulation. The concentration of holes is assumed to be much larger than the concentration of electron traps, and an electron can tunnel only to its nearest hole.

Recently, Kitis and Pagonis [27] obtained analytical solutions for the set of differential equations in the model of Jain et al. [26] by using certain mathematical and physical simplifications. These authors presented analytical expressions describing thermally and optically stimulated luminescence signals within this model. In the case of isothermal TL decay the corresponding analytical expression is:

$$I(t) = \frac{3 n_o \rho' z F(t)^2}{1 + z t/\tau} \cdot exp(-\rho' F(t)^3) \quad (2)$$

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

$$\tau = s^{-1}\, e^{\frac{E}{kT_{dec}}} \quad (4)$$

where $n_0$ the initial concentration of trapped electrons, $z$ a constant, $E$ the activation energy, $s$ the frequency factor, $\tau$ the lifetime, $\rho'$ a dimensionless parameter representing the normalized donor–acceptor density and $T_{dec}$ the temperature at which the isothermal TL experiment is carried out. Within this localized transition model (as well as in delocalized TL models), the lifetime $\tau$ in Eq. 4 should depend very strongly on the PID temperature $T_{dec}$.

It is worth to mention here that an isothermal decay expression based on a tunneling pair recombination model is given by Chang and Thioulouse [28]. The final expression of Chang and Thioulouse [28] (Eq. 12 in their article) is not completely analytical so, it cannot be used for direct fitting of experimental data. However, qualitatively has the same meaning with Eq. 2, i.e., explains why the isothermal decay does not follow exactly the $t^{-1}$ low, but depends also on the donor–acceptor distribution established by the irradiation.

In this section the experimental PID curves are fitted with a sum of a tunneling component given by Eq. 2, plus a single exponential component. Examples of this type of analysis, using a tunneling plus an exponential component, are given in Fig. 5, with the isothermal signal shown normalized at the intensity at time $t = 0$. Figs. 5(a), (b) show results for "zero storage" samples, whereas Figs. 5(c), (d) show those for "24 h storage" samples. The parameter values obtained from this type of analysis are listed in Table 1. The values of lifetime $\tau$ for "zero storage" and "24 h storage" samples were identical, $0.924 \pm 0.012$ and $0.995 \pm 0.020$, correspondingly. The FOM values were from 2.5% at $T_{dec}$ 160 °C to 1%

at $T_{dec}$ 250 °C. The principal observations from this analysis can be summarized as follows:

- The lifetime $\tau$ was found to be the same for all temperatures used in the isothermal TL experiment, and also the same for "zero" and "24 h storage" samples. This result contradicts the expected behavior described by Eq. 4.
- Similarly, the value of the dimensionless parameter $\rho'$ was found to be the same for all temperatures used in the isothermal TL experiment, and also the same for "zero" and "24 h storage" samples. This result is in agreement with the physical assumptions of the model by Jain et al. [26].
- The contribution of the tunneling component to the total decay curve in the case of "24 h storage" samples is slightly smaller than the contribution of the same component for the "zero storage" sample. This result makes physical sense since the tunneling component can be expected to have decayed significantly within the 24 h storage period.
- The behavior of the exponential component is similar to the expected behavior from delocalized TL models. This is shown by the Arrhenius plot of $ln(\tau)$ versus $1/kT_{dec}$ which is shown in Fig. 6. The values of the activation energies obtained from this figure are 0.70 eV and 0.77 eV for the "zero storage" and "24 h storage" samples correspondingly.

Based on the above observations, it is concluded that the analysis of the PID curves based on a tunneling component plus an exponential component provides a partial explanation of the experimental results, except for the fact that the $\tau$ values are independent of the temperatures used in the isothermal experiments.

### 3.3. Analysis based on tunneling component

In this section the experimental PID curves are fitted using a single tunneling component given by Eq. 2 plus a stable background. This type of analysis gave the best fits among all fitting

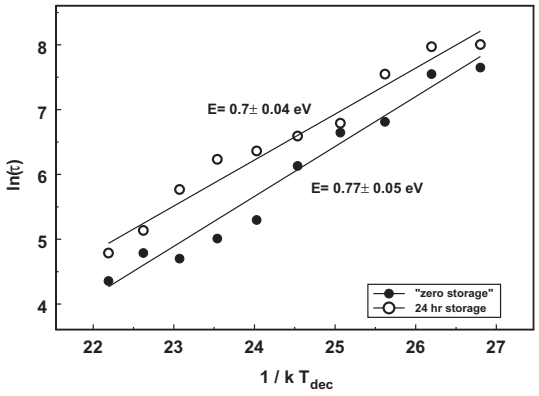


**Fig. 6.** Activation energies of the exponential component obtained from the analysis using the sum of tunneling plus and exponential components in Section 3.2.

procedures presented in this paper since the FOM values for $T_{dec}$ above 200 °C drops down to 1%. Typical examples are shown in Fig. 7 with (a) the PID curve at 240 °C for "zero storage" sample, (b) the PID curve for "24 h storage" samples and (c) the constant background component. The specific conclusions from this analysis were:

- The values of $\tau$ given in Table 1, were $1.44 \pm 0.14$ s and $1.12 \pm 0.3$ s for "zero storage" and "24 h storage" cases correspondingly. These values are in reasonable agreement with the values of the tunneling component in the previous analysis listed in Table 1.
- Once more, the constancy of the values of $\tau$ at different temperatures is in disagreement with the expected behavior from Eq. 4.
- The value of $\rho'$ was not constant, but increased with the isothermal temperature $T_{dec}$, as shown in Fig. 8. This is in disagreement with the model of Jain et al. [26], which requires a constant value of $\rho'$.

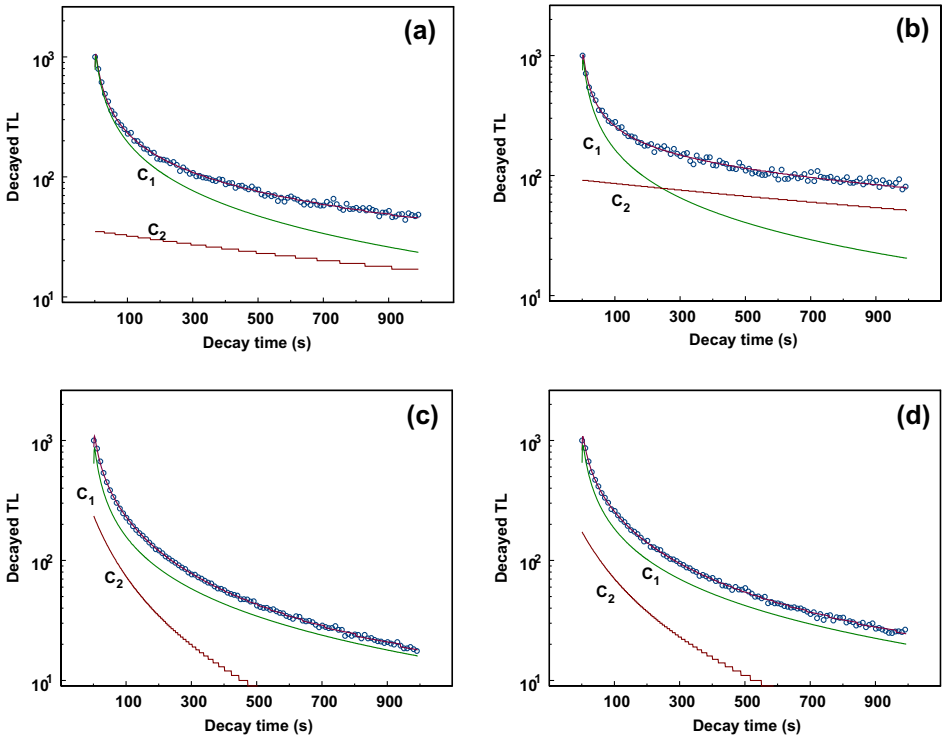


**Fig. 5.** Examples of PID curves using a sum of a tunneling plus an exponential component. (a) PID at 170 °C for "zero" storage time. (b) PID at 170 °C for 24 h storage time. (c) PID at 240 °C for "zero" storage time. (d) PID at 240 °C for 24 h storage time.

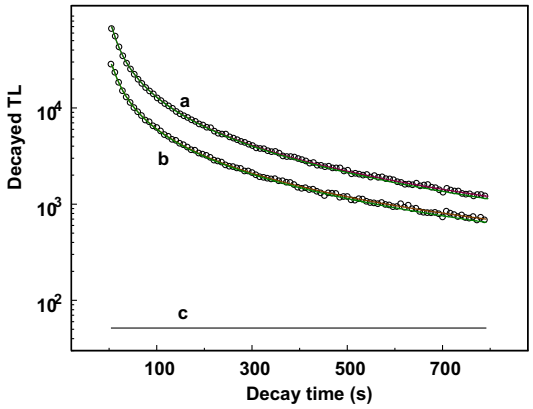


**Fig. 7.** PID curves fitted by a single tunneling component. $T_d$ = 240 °C (a) "zero", (b) 24 h storage time, (c) the level of the stable component.

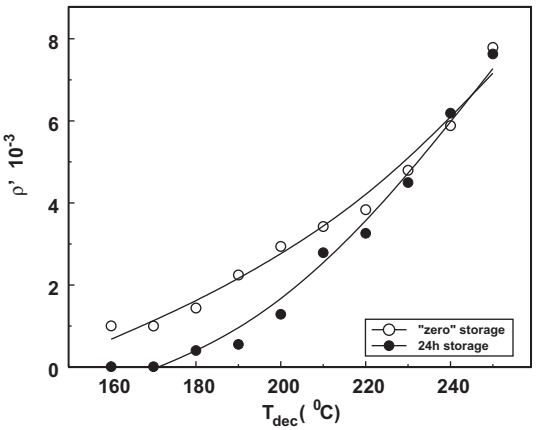


**Fig. 8.** The parameter $\rho'$ for the one tunneling component analysis of PID curves in Section 3.3.

The high quality of the fits to the experimental data using the single tunneling component is a very interesting result. Unfortunately, the results for both $\tau$ and $\rho'$ are not in principle compatible with the tunneling model of Jain et al. [26]. However, a more detailed discussion in the next section provides a possible explanation for the observed behaviors of both $\tau$ and $\rho'$.

## 4. Discussion

As discussed in the previous sections, the behavior of the experimental PID curves of Durango apatite is in disagreement with the behavior expected from conventional TL kinetic models based on delocalized transitions. The excellent fits to experimental curves obtained in all types of analysis presented in this paper do not allow a unique answer about which model gives the correct physical description. The main result that must be discussed and explained is the constancy of the lifetime $\tau$ at different isothermal temperatures. Furthermore, the dependence of the parameter $\rho'$ on the isothermal temperature shown in Fig. 8 needs to be discussed and explained.

The mean lifetime $\tau$ is a kinetic quantity given by Eq. 4 and varies strongly with $T_{dec}$. However, the experimental results show that $\tau$ is nearly independent on $T_{dec}$. One possible explanation for this disagreement is the following.

According to Eq. 4, $\tau$ depends on both $s$ and $E$. A stable value of $\tau$ could mean that either $s$ or $E$ depend on the isothermal temperature $T_{dec}$. By rearranging Eq. 4 one obtains:

$$E(T) = ln(\tau \cdot s) \cdot k \cdot T_{dec}. \quad (5)$$

In the model by Jain et al. [26], the temperature activates the electrons to a higher excited energy level, from which they can either recombine or be retrapped into the ground state. In this model the frequency factor $s$ and the retrapping probability are equal. From a physical point of view, one might expect that as $T_{dec}$ increases, electrons may be activated to a *progressively* higher energy level state, especially if a continuum of energy states is available near the excited state. The possibility of such a continuum of states within the energy gap in feldspars has been considered in detail in the work by Poolton et al. [9,10] and in the recent extensive work by Jain and Ankjrgaard [29]. If one assumes for example a constant $s$ value of $10^{13}$s, the known values of $\tau$ from Table 1, and temperatures $T_{dec}$ between 150 and 250 °C, one can evaluate the expected values of activation energy $E$ by using Eq. 5.

The results of these calculations are shown in Fig. 9, where a rather small increase of the energy E by $\Delta E \simeq 0.2$ eV can explain the constant value $\tau$. According to this argument, the experimentally observed constant lifetime $\tau$ at increased temperatures $T_{dec}$, can be explained in the framework of the model by Jain et al. [26] by the fact that the electron recombination takes place each time from a progressively higher excited energy level, which differs by a small amount of $\Delta E$. The total energy difference $\Delta E$ required between the temperatures $T_{dec}$ of 150 and 250 °C, is only 0.2 eV.

From a physical point of view, the value of the fitting parameter E possibly represents an effective energy which describes access to available higher energy levels as the temperature of the isothermal experiment is increased. Another possible physical origin of this energy variation could be phonon vibrations, or the expected lattice disorder in feldspars [9,10].

Our explanation for the observed constant lifetime is based on the assumption of a constant frequency factor s, and on a variable effective energy $E$. Another possible explanation for this almost constant value of the lifetime is its strong dependence on the trap-center distances. As discussed in the theoretical work by Poolton et al. [9,10], one can expect significant disorder in the crystalline structure of these materials, due to the presence of a distribution of depths and widths in adjacent quantum wells. Such a distribution of distances and energies could in principle lead to an "averaging" of the observed lifetimes at different temperatures, as observed in our experiment. Based on the data available, it is not possible to decide on the exact origin of the constant lifetime, and further experimental work is required to decide between these two explanations.

The increase of the activation energy as $T_{dec}$ increases can also explain the observed increase of $\rho'$. In the model by Jain et al. [26] the extent and the overlap of the electron wave functions within the

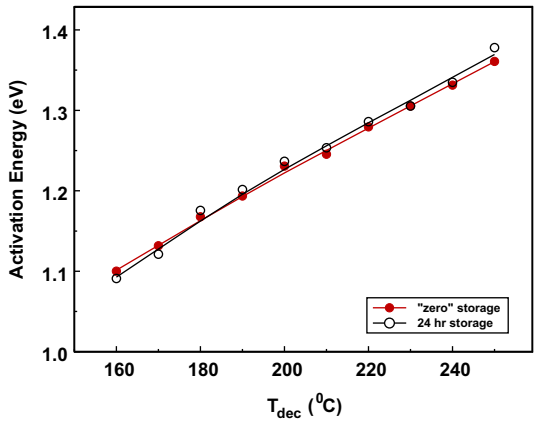


**Fig. 9.** Estimated activation energy as a function of $T_{dec}$, as evaluated from Eq. 5.

donor–acceptor system and for adjacent electron hole pairs is of crucial importance for determining the electron tunneling recombination. As the isothermal temperature $T_{dec}$ increases, the extent and the overlap of the electron wave functions increases, and this leads also to an increase of tunneling recombination probability. This results in turn to effective higher donor–acceptor distances, a fact which is expressed as an increased value of $\rho'$.

Although the single tunneling component analysis seems to have some advantages over the tunneling plus conventional exponential component analysis, in our opinion, a final decision for which analysis is correct can be made only if the isothermal TL decay experiments were carried out on samples possessing a natural TL signal. The reason is that the tunneling component is expected to be drastically reduced during geological times, so that the conventional exponential component will be dominant in the Natural TL glow-curve.

This effect of a dominant single exponential component should also be evident in the case of the "24 h storage samples" studied in this paper. The signal loss due to tunneling is estimated in the present work by dividing the integrated TL signal under each PID curve for the "24 h storage time" samples by the corresponding integrated signal for "zero storage" samples. The precent signal loss factor evaluated with this method is found to be $0.545 \pm 0.044$ in excellent agreement with previous results [16]. However, the experiments described in this paper did not find any evidence of a dominant single exponential component in the "24 h storage" samples. In conclusion, it is necessary to carry out further experimental work on samples possessing a natural TL signal, in order to clarify which of the two models (single tunneling component vs tunneling plus exponential components) describes better the physical situation.

## 5. Conclusions–implications

The isothermal TL decay of Durango apatite does not obey conventional TL kinetic theory. The TL decay constant $\lambda = 1/\tau$ was found to be independent of temperature, instead of the strong temperature dependence expected from conventional delocalized models. The isothermal decay results presented in this work can be considered a direct observation of radiative tunneling recombination processes in this material. The isothermal TL curves are described excellently by new analytical expressions [27] derived within the newly developed tunneling kinetic model of Jain et al. [26].

## References


[1] A.G. Wintle, Nature 245 (1993) 143.
[2] A.G. Wintle, J. Lumin. 15 (1977) 385.
[3] R. Visocekas, T. Ceva, C. Marti, F. Lafaucheux, M.C. Roberts, Phys. Stat. Sol. (a) 35 (1976) 315.
[4] R. Visocekas, A. Geoffry, Phys. Stat. Sol (a) 41 (1977) 4993.
[5] R. Visocekas, Nucl. Tracks 10 (1985) 521.
[6] R.H. Templer, Radiat. Prot. Dosim. 17 (1986) 493.
[7] S. Tyler, S.W.S. McKeever, Nucl. Tracks Radiat. Meas. 14 (1986) 149.
[8] R. Chen, Radiat. Meas. 32 (2000) 505.
[9] N.R.J. Poolton, J. Wallinga, A.S. Murray, E. Bulur, L. Bøtter-Jensen, Phys. Chem. Miner. 29 (2002) 210.
[10] N.R.J. Poolton, K.B. Ozanyan, J. Wallinga, A.S. Murray, L. Bøtter-Jensen, Phys. Chem. Miner. 29 (2002) 217.
[11] N.R.J. Poolton, R.H. Kars, J. Wallinga, A.J.J. Bos, J. Phys. Cond. Matt. 21 (2009). Art. no. 485505.
[12] B. Li, S.H. Li, J. Phys. D: Appl. Phys. 41 (2008) 22. Art. no. 225502.
[13] A. Larsen, S. Greilich, M. Jain, A.S. Murray, Radiat. Meas. 44 (2009) 467.
[14] G. Kitis, P. Bousbouras, C. Antypas, S. Charalambous, Nucl. Tracks Radiat, Meas. 18 (1991) 61.
[15] G. Kitis, G.S. Polymeris, V. Pagonis, N.C. Tsirliganis, Phys. Stat. Sol. (a) 203 (2006) 3816.
[16] G.S. Polymeris, N. Tsirliganis, Z. Loukou, G. Kits, Phys. Stat. Sol. (a) 203 (2006) 578.
[17] N. Tsirliganis, G.S. Polymeris, Z. Loukou, G. Kitis, Radiat. Meas. 41 (2006) 954.
[18] N. Tsirliganis, G.S. Polymeris, G. Kitis, V. Pagonis, J. Lumin. 126 (2007) 303.
[19] R. Visocekas, Radiat. Prot. Dosim. 100 (2002) 45.
[20] M.R. Baril, Spectral investigations of luminescence in feldspars (Ph.D. thesis), Simon Fraser University 2002, Burnaby, BC, Canada. Available online at: <www.cfht.hawaii.edu/wbaril/Temp/barilphdthesis.pdf>.
[21] M.R. Baril, D.J. Huntley, J. Phys. Cond. Matt. 15 (2003) 8011.
[22] R. Chen, S.W.S. McKeever, Theory of Thermoluminescence and Related Phenomena, World Scientific, Singapore, 1997.
[23] C. Furetta, G. Kitis, J. Mater. Sci. 39 (2004) 2277.
[24] Balian, Eddy, Nucl. Instr. Methods 145 (1977) 389.
[25] Y. Kirsh, R. Chen, Nucl. Tracks Radiat. Meas. 18 (1991) 37.
[26] M. Jain, B. Guralnik, M.T. Andersen, J. Phys.: Condens. Matter 24 (2012) 385402.
[27] G. Kitis, V. Pagonis, J. Lumin. 137 (2013) 109.
[28] I.F. Chang, P. Thioulouse, J. Appl. Phys. 53 (1982) 5873.
[29] M. Jain, C. Ankjrgaard, Radiat. Meas. 46 (2011) 292.