Determine dose-saturation level from thermoluminescence curves by the GOK and OTOR models

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ABSTRACT
This study shows that the pre-exponential frequency in the general-orders of kinetics (GOK) model depends on the ratio between the initial number of trapped electrons ($n_0$) and the total concentration of trapping states ($N$) as well as the frequency factor. The frequency factor can be calculated by the one trap-one recombination (OTOR) model and does not depend on the ratio of $n_0/N$. That means the ratio of $n_0/N$ can be deduced based on a comparison of the GOK and OTOR models. When the ratio of $n_0/N$ is approximately 1, the sample is in the dose-saturation level. Therefore, at the first time, this study has determined the dose-saturation level of the sample from the thermoluminescence (TL) curve fitting.

1. Introduction
To check the dose-saturation level of the irradiated samples, the samples need to be irradiated many times with gradually increasing doses until the samples reach the dose-saturation level. At each time of irradiation, the samples should be checked if the thermoluminescence (TL) signal strength does not increase further, the samples reach saturation. TL spectrometry measurements should be performed quickly and accurately to ensure that the TL signal is not blurred or reduced by the fading effect. The experimental measurements to test the saturation can be carried out but take a lot of samples and take much time to complete. Therefore, to test the samples’ saturation, a semi-empirical method has been shown based on the fitted curves of TL by the general-orders of kinetics (GOK) model [1] and one trap-one recombination (OTOR) model [2,3]. When the figure of merit of two models is relatively small that allows us to compute the dynamic parameters of the TL spectrum by GOK and OTOR models. The kinetic parameters of TL glow peaks of chilli powder irradiated by gamma rays with the different doses of 0, 4 and 8 kGy have been calculated and estimate by computerized glow curve deconvolution method and the R package tgcd by using the TL glow curve data [4]. From these parameters, calculating the saturation ratio between the initial number of trapped electrons ($n_0$) and the total concentration of trapping states ($N$) are presented. The samples reach the dose-saturation level when the ratio between $n_0$ and $N$ is much different from 1 [5,6]. Thus, based on the TL curves of the samples, the saturation of the samples that brings to the conclusions about the respective dose-saturation level was determined.

The first work this study needs to do is simulating the TL curves of the initial parameters given in the kinematic models. Simulated models may have one or more peaks, which may be saturated or unsaturated. For experimental samples, The spectrum from the GLOCANIN project [7,8] is used. This empirical TL curve is the overlap of many complicated TL peaks that were conducted to separate peaks each other and determine the saturation values of each peak based on this study results. In addition, the TL curves of irradiated pepper samples with 8 kGy dose with 360 and 720 h [4] after irradiation were also included to determine dose-saturation level.

2. Material and methods
2.1. Simulation of TL curves
Both saturated and unsaturated samples were simulated and computed basing the R package tgcd [4,9]. The simulated samples include the saturated samples with a spectral peak (TL1), the unsaturated samples with a spectral peak (TL2), the saturated samples with two spectral peaks (TL3) and the unsaturated samples with two spectral peaks (TL4). The parameters for the sample simulation are the energy trap values ($E$), heating rate ($\beta$), the frequency factor ($s$), electron trapping factor.
Table 1. Parameters for simulating the TL curves of samples.

| Peaks | n₀   | N   | E (eV) | β (K/s) | s (−1) | A₀   | A₉   |
|-------|------|-----|--------|---------|--------|------|------|
| TL1   | 10⁸  | 10¹⁰| 0.56   | 1       | 10¹⁴   | 10⁻³ | 10⁻⁷ |
| TL2   | 10⁸  | 10¹⁰| 0.56   | 1       | 10¹⁴   | 10⁻³ | 10⁻⁷ |
| TL3   | 10⁹  | 10¹⁰| 1.50   | 1       | 10¹⁴   | 10⁻³ | 10⁻⁷ |
| TL4   | 10⁹  | 10¹⁰| 1.68   | 1       | 10¹⁴   | 10⁻³ | 10⁻⁷ |

(A₀) and in hole trap coefficient (A₉). The initial number of trapped electrons (n₀) and the total concentration of trapping states (N) are shown in Table 1. These parameters are simulated by the simqOTOR function of the R package tgc by the OTOR model [2,3,9]. These TL1 and TL3 samples are the two TL spectra that are assigned with the dose-saturation level. Therefore, the initial number of trapped electrons and the total concentration of trapping states of the two samples TL1 and TL3 are made equal to each other. For unsaturated samples, TL2 and TL4, the initial number of trapped electrons and the total concentration of trapping states are made different from each other.

2.2. The TL curves obtained from experiments

The experimental curves used are the complex TL curves with the overlap of many single peaks. The data are referenced by the GLOCANIN project [7,8]. This project has been widely used in the TL researches [3,9,10]. However, the determination of saturation of these two curves Refglow002 and Refglow009 has not been mentioned yet. Among them, the TL curves with the overlap of many single peaks are still unknown about the dose-saturation level. In this paper, the two TL curves of Refglow002 and Refglow009 of the project GLOCANIN are used to calculate the individual dose-saturation level for each peak.

In addition, the two TL curves of the chilli powder sample irradiated at a dose of 8 kGy and preserved 360 h and 720 h after being irradiated were used to determine saturation. TL curves of the chilli samples fitted to both the GOK and OTOR models are mentioned [4]. The chilli powder samples irradiated with a dose of 8 kGy were tested for their saturation.

2.3. Methods

The calculation basis of the method is based on the kinetic parameters such as activation energy (E), the order of kinetics (β), trapping and recombination probability coefficients (R), frequency factors (s), the equation for determining the value of s (GOK model) that are given by [11]:

\[ s = \left( \frac{\beta E}{kT_M^2} \right) \exp \left( \frac{E}{kT_M} \right) \left[ 1 + (b - 1) \times \frac{2kT_M}{E} \right]^{-1} \]  

Using the equation of the pre-exponential frequency s”, applied to the OTOR model [2,11]:

\[ s'' = \left( \frac{N}{n_0} \right)^{b-1} \left( \frac{\beta E}{kT_M^2} \right) \exp \left( \frac{E}{kT_M} \right) \times \left[ 1 + (b - 1) \times \frac{2kT_M}{E} \right]^{-1} \]  

Thus, the relation between s” and s is:

\[ s'' = s \left( \frac{n_0}{N} \right)^{b-1} \]  

The value of s (of OTOR model) is computed [2,3] by the equation:

\[ s = \left( \frac{\beta E}{kT_M^2} \right) \exp \left( \frac{E}{kT_M} \right) \left[ 1 + W[\exp(Zm)]^2 \right] \left[ 1 + 2W[\exp(Zm)] \right] (1 - R) \]  

The calculation equation of the value of s in the OTOR model absolutely bases on the physical model [2,3] whereas the GOK model bases on the semi-empirical condition.

The calculating condition of the GOK and OTOR models produces very small errors, then we can rely on E, b, R to compute (n₀/N)b−¹ by the equation:

\[ \left( \frac{n_0}{N} \right)^{b-1} = \frac{s''}{s} = \left[ 1 + (b - 1) \times \frac{2kT_M}{E} \right] \times \left[ 1 + W[\exp(Zm)]^2 \right] \left[ 1 + 2W[\exp(Zm)] \right] (1 - R) \]  

The analysis of the TL curve according to the GOK model is given by:

\[ I(T) = I_M b^{\frac{E}{kT}} \exp \left( \frac{E}{kT_M} \right) \times \left[ \frac{T - T_M}{T_M} \right] \times \left[ (b - 1) \times \frac{T^2}{kT_M^2} \exp \left( \frac{E}{kT_M} \right) \times \frac{T - T_M}{T_M} \right] \times \left[ \left( 1 - \frac{2kT}{E} \right) + 1 + (b - 1) \times \frac{2kT_M}{E} \right]^{-\frac{1}{kT}} \]  

The descriptions of the OTOR model are presented in [2]. The analysis of the TL curve according to the OTOR model is based on the Lambert W function [2,3,12].
as follows:

$$I(T) = I_M \frac{W[\exp(Z)] + W[\exp(ZmM)]^2}{W[\exp(Z)] + W[\exp(Zm)]^2} \exp \left( \frac{E}{kT} \times \frac{T_M - T}{T_M} \right)$$

$$Zm = \frac{R}{1 - R} - \ln \left( \frac{1 - R}{R} \right) + E \frac{kT_M^2}{kT} (1 - 1.05^{1.26}) F(T_M, E)$$

$$Z = \frac{R}{1 - R} - \ln \left( \frac{1 - R}{R} \right) + E \frac{kT_M^2}{kT} (1 - 1.05^{1.26}) F(T, E)$$

$$F(T_M, E) = T_M \exp \left( -\frac{E}{kT_M} \right) + \frac{E}{k} E_i \left( -\frac{E}{kT_M} \right)$$

$$F(T, E) = T \exp \left( -\frac{E}{kT} \right) + \frac{E}{k} E_i \left( -\frac{E}{kT} \right)$$

Among them, $W(x)$ is the Wright Omega function of the variable $x$, $E_i$ is the exponential integral function [12], $R = R_0/A_m$, $A_n$ and $A_m$ are in turn the trapping probability and recombination, $R$ in OTOR model is equivalent with the kinetic order b of GOK model. The Wright Omega function suggested by Singh and Gartia [13–15] aims to replace the Lambert $W$ function given by Kitis and Vlachos [2,3,12]. Singh and Gartia showed the advantage of the Wright Omega function in comparison with the Lambert $W$ function [14], as pointed out by Lawrence [16].

After fitting the glow curves by the GOK model and OTOR model, the ratio $n_0/N$ can be determined to check the dose-saturation level of the sample. The TL glow curves fitting depends on the figure of merit (FOM) given by:

$$\text{FOM} = \frac{\sum_p |y_{\text{exp}} - y_{\text{fit}}|}{\sum_p y_{\text{fit}}}$$

where $y_{\text{exp}}$ and $y_{\text{fit}}$ are the experimental data and the values of the fitting function, respectively.

### 3. Results and discussion

Calculation results of saturated and unsaturated samples of the simulated and empirical samples, including single peak or multiple overlapping peaks are based on the R package tgcd [9]. The formula (6) into the R package tgcd to calculate the dose-saturation level of the simulated and experimental samples have been inserted. Here, the results obtained have many kinetic parameters but they only showed the ratio $n_0/N$ and the figure of merit (FOM) because it relates to the determination of the dose-saturation level behaviour of the simulated and experimental samples.

The results showed that the $n_0/N$ ratio of TL1, TL2 samples were $0.99 \pm 0.01$ and $0.32 \pm 0.05$, respectively. The TL glow curves of TL1 and TL2 are given by Figure 1. Toward samples with two peaks, the ratio $n_0/N$ of the first and second peaks has values of $0.99 \pm 0.03$ and $0.99 \pm 0.02$ for TL3; $0.11 \pm 0.05$ and $0.12 \pm 0.02$ for TL4. Thus, with the glow curve of TL1 the $n_0/N$ ratio is close to 1 and thus the sample of TL1 is saturated. With the TL2 pattern, the ratio $n_0/N$ is not equal to 1 and hence the sample of TL2 is not saturated. The results for the glow curves of TL1 and TL2 are consistent with the initial simulation. However, with the glow curve of TL2 unsaturated level, the value $n_0/N$ between simulation and computation is very different from each other. For the glow curve of TL3 simulation, they were shown both peaks dose-saturation level. Calculation results indicate that it is suitable for simulation. The calculation and fitting the glow curves of TL3 and TL4 consists of two.

![Figure 1. Fitting TL glow curve to determine dose-saturation level of TL1 and TL2.](image-url)
peaks given in Figure 2. For the fitting the glow curve of TL4, they were showed both peaks unsaturated. The results of calculation also show that both peaks of TL4 were not saturated.

Toward the glow curve fitting Refglow002, it consists of four main peaks. Results showed that the scores for \( n_0/N \) of the four peaks were determined as 0.99 ± 0.01, 0.99 ± 0.06, 0.99 ± 0.03 and 0.99 ± 0.01, respectively. These peaks give the ratio of \( n_0/N \) to nearly 1. Therefore, all four peaks of Refglow002 were nearly saturated. For the glow curve of Refglow009, it consists of nine main peaks. The results show that the scores for \( n_0/N \) of the nine peaks were determined as 0.14 ± 0.07, 0.63 ± 0.05, 0.02 ± 0.09, 0.13 ± 0.39, 0.01 ± 0.07, 0.20 ± 0.38, 0.19 ± 0.53, 0.88 ± 0.45 and 0.02 ± 0.06. These peaks give the ratio \( n_0/N \) very different from the value 1. Therefore, the peaks of the current Refglow009 sample have not yet reached the dose-saturation level. The computation of the glow curve of Refglow002 and Refglow009 samples consisted of multiple overlapping peaks given by Figure 3.

Toward two samples of chilli powder irradiated at a dose of 8 kGy preserved 360 h and 720 h after being irradiated, their glow curves showed only peak single. Results showed that the \( n_0/N \) ratio of the peak single of the two samples of chilli powder irradiated with the 8 kGy radiation dose stored after 360 and 720 h irradiation was determined to be 0.28 ± 0.08 and 0.25 ± 0.04. Therefore, the sample of chilli powder with the current 8 kGy irradiation dose has not yet saturated. The glow curves of chilli powder were given by Figure 4.

The results for the dose-saturation level determination of the sample are shown in Tables 2 and Table 3. With the glow curves of TL1, TL2, TL3 and TL4, the calculated results show that the FOM of the OTOR has less value than the FOM of the GOK model. Therefore, the simulation spectrum is more consistent with the OTOR model than the GOK model. This result is appropriate because when simulated by using the OTOR model and

![Figure 2. Fitting TL glow curve to determine dose-saturation level of TL3 and TL4.](image)

![Figure 3. Fitting TL glow curve to determine dose-saturation level of Refglow002 và Refglow009.](image)
Figure 4. Fitting TL glow curve to determine dose-saturation level of chilli powder sample irradiated with 8 kGy (stored 360 h and 720 h).

Table 2. The results determine the saturation of the TL curves.

| TL curves | FOM (OTOR) | FOM (GOK) | Peaks | n₀/N   | Result     |
|-----------|------------|-----------|-------|--------|------------|
| TL1       | 0.001      | 0.012     | P1    | 0.99 ± 0.01 | Saturation |
| TL2       | 0.007      | 0.028     | P1    | 0.32 ± 0.05  | Unsaturation |
| TL3       | 0.001      | 0.011     | P1    | 0.99 ± 0.03  | Saturation   |
| TL4       | 0.002      | 0.071     | P1    | 0.11 ± 0.05  | Unsaturation |
| Refglow002| 0.009      | 0.009     | P1    | 0.99 ± 0.01  | Saturation   |
| Refglow009| 0.077      | 0.078     | P1    | 0.14 ± 0.07  | Unsaturation |

8 kGy (360 h) 0.329 0.346 P1 0.83 ± 0.08 Unsaturation
8 kGy (720 h) 0.322 0.357 P1 0.52 ± 0.04 Unsaturation

Table 3. The results determine the saturation of experimental samples (chili powder stored after 360 h).

| TL curves | FOM (OTOR) | FOM (GOK) | E (eV) | n₀/N   | Result     |
|-----------|------------|-----------|-------|--------|------------|
| 0 kGy     | 0.92       | 0.91      | 0.95  | 0.45 ± 0.02 | Unsaturation |
| 2 kGy     | 0.68       | 0.67      | 0.96  | 0.51 ± 0.05 | Unsaturation |
| 4 kGy     | 0.51       | 0.49      | 0.98  | 0.62 ± 0.03 | Unsaturation |
| 6 kGy     | 0.38       | 0.37      | 0.99  | 0.85 ± 0.03 | Unsaturation |
| 8 kGy     | 0.32       | 0.34      | 0.98  | 0.91 ± 0.08 | Unsaturation |
| 10 kGy    | 0.75       | 0.76      | 1.03  | 0.95 ± 0.08 | Unsaturation |

4. Conclusions

The article presents the method of calculating the dose-saturation level of simulated and experimental samples including the peaks of glow curves of TL. Simulation and computation are performed based on the R package textcom. The results of the calculation are consistent with simulation and experiment. The method of determining the dose-saturation level of the sample by comparing the kinetics parameters between the two GOK and OTOR models is first investigated and applied. The method is more suitable if the coefficient of joints FOM is small. The results show that the peaks of the Refglow002 sample are nearly saturated, while the peaks of Refglow009 have not reached dose-saturation level. With the sample of chili powder irradiated at 2, 4, 6, 8 and 10 kGy dose, the sample has not reached the dose-saturation level. Accurate determination of calculation of all samples is, the smaller the FOM of the two models is.

The saturation determination of the simulated and experimental samples basing on the glow curves of TL was first investigated. The calculation results depend on the FOM’s fitting for both the GOK and OTOR models. For the spectral peak of the unsaturated sample, the ratio n₀/N between the simulation and the calculation of the unsaturated peaks varies greatly. The reason for the difference is individual errors that lead to total errors during the calculation and fitting of both models. Therefore, in the calculation process, if the errors of fitting are too grave, this method cannot be applied to determine the dose-saturation level of the sample. In this study, the simulated and calculated spectra brought small values to FOM values for both the GOK and OTOR models. Therefore, it is possible to apply this study to determine the dose-saturation level from the glow curves of TL.
sample saturation will be the basis for studying applications related to dose measurement, TL material preparation, detection and identification of irradiated food.

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