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# Preliminary Thermoluminescent Dosimeter Glow Curve Analysis with Automated Glow Peak Identification for LiF:Mg,Ti

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**Abstract**—When appropriately analyzed, thermoluminescent dosimeter glow curve analysis allows for improved quantification of thermoluminescent material behavior while flagging abnormalities. The mathematical separation of a glow curve into contributions from energetically unique trap states, or glow curve analysis, may be used to remove undesired effects of signal fading for complex materials. A generalized glow curve analysis software for the separation of glow curves is presented in this paper. Written in C++, the software uses the first-order kinetics model with automatic peak identification. The automatic identification of peaks is achieved through a unique peak-finding algorithm. The program was performance tested using experimental glow curve data from LiF:Mg,Ti, and comparative results are presented. *Health Phys.* 120(00):000–000; 2021

**Key words:** analysis, statistical; dosimetry, thermoluminescent; kinetics; quality assurance

## INTRODUCTION

THERMOLUMINESCENT DOSIMETERS (TLDs) are broadly used for environmental and external personnel radiation dosimetry. The thermoluminescence (TL) properties of many materials are known. Analysis of the behavior of TL material benefits from the separation of individual glow curves of the emitted TL spectra called glow curve analysis (GCA). Glow curve analysis is also useful for quality control, neutron-gamma dosimetry, and surface dose estimation (Horowitz and Moscovitch 1986; Horowitz and Yossian 1995; Basun et al. 2003).

While the exact mathematical function of TL spectra is a subject of debate, many GCA programs have been written using the first-order kinetics model (Moscovitch et al. 1983;

Lilley and McKeever 2000). The first-order kinetics model is often fit to experimental data using the Levenberg-Marquardt algorithm (LMA) (Kitis et al. 1998; Harvey et al. 2011). The earliest GCA codes were developed using programming languages, but more recent work employed mathematics parsing platforms such as MATLAB. Mathematics parsing platforms are difficult to integrate into portable embedded systems. Most importantly, the overwhelming majority of GCA programs require the manual input of initial peak-fitting parameters. This approach is susceptible to convergence failures and errors, turning the process of creating accurate glow curves into more of an art form than a science.

This paper presents a new Glow Curve Analysis (GCA) software that automates the identification and fitting of individual glow peaks for TLD eliminating human intervention. The code is based on the first-order kinetics model because of its simplicity and universal application. The fitting is implemented using the Levenberg-Marquardt algorithm because of its stability. The program's performance on a variety of TLD input is presented. Additionally, the GCA software in this paper uses only C++ and its standard libraries, ensuring portability.

## METHODS AND MATERIALS

### Physics methods

**First-order kinetics model.** Models explaining the nature of TL glow curves have been developed with parameters determined using experimental data for many TLD materials. A first-order kinetics (FOK) model for TL glow curves is (Kitis et al. 1998):

$$I(T) = I_m \exp \left[ 1 + \frac{E}{KT} \frac{T-T_M}{T_M} - \frac{T^2}{T_M^2} \times \exp \left( \frac{E}{KT} \frac{T-T_M}{T_M} \right) (1-\Delta) - \Delta_m \right], \quad (1)$$

where  $I(T)$  is the peak intensity  $I$  at temperature  $T$  in K,  $I_m$  is the intensity at the peak maximum,  $E$  is the activation energy in eV,  $k$  is Boltzmann constant in eV K<sup>-1</sup>, and  $T_m$  is the temperature at the peak maximum in K,  $\Delta$  is  $2kT(E)^{-1}$ , and  $\Delta_m$  is  $2kT_m(E)^{-1}$ .

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**Activation energy.** The activation energy,  $E$ , is approximated as (Chen 1969):

$$E = C_{\tau} \frac{kT_m^2}{\tau} - b (2kT_m), \quad (2)$$

where  $C_{\tau}$  is  $1.5 + [3 \times (m_g) - 0.42]$ ;  $b$  is  $1.48 + [4.2 \times (m_g) - 0.42]$ ;  $m_g$  is  $\frac{w_l}{w_r}$ , where  $w_l$  and  $w_r$  are the half-width at the low temperature side of the peak; and the half-width toward the fall of the glow peak, respectively,  $\tau$  is  $T_2 - T_1$ . The first-order

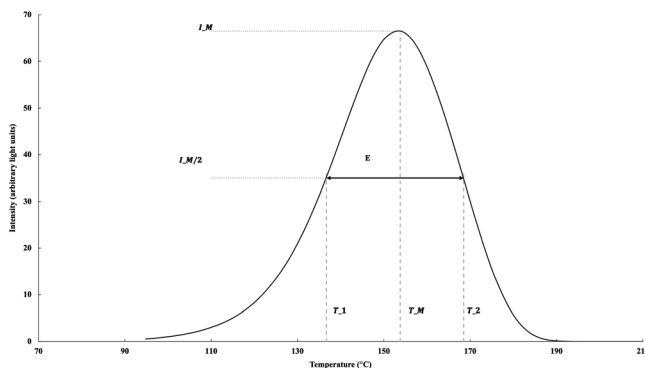
**F1** kinetics model parameters are shown explicitly in Fig. 1.

## Software methods

**Overview.** The GCA software in this paper is written for C/C++17 (ISO Standard 2017). C++ was chosen for its quick processing speeds, cross-platform compatibility, and lightweight distribution. This software utilizes the Standard Template Library (STL). The software accepts a directory of comma-separated values (CSV) files as input. This is a popular format for TLD readers, and file conversion to CSV is easily achieved. The software consists of a five-stage process: batch file handling, data noise reduction, automated peak detection, curve fitting, and output. This process flow is visualized in Fig. 2. Additionally, the software offers the user the option to input initial peak values, which will override the automated peak detection portion of the code.

**F2**

**Time complexity.** In this paper, the time complexity (sometimes called the “complexity”) of various algorithms will be given approximately. The time complexity of an algorithm or program is defined as how many operations are performed before completion, sometimes as a function of the input. Thus, the time complexity is an effective method of measuring the speed of a function or of a program as a whole (Cobham 1965).



**Fig. 1.** The parameters of the first-order kinetics model.  $I_M$  is the maximum intensity of the glow peak, and  $T_M$  is the temperature at which it occurs.  $T_1$  is the temperature where the left full-width half maximum of the glow curve is found, and  $T_2$  is the temperature at which the right full-width half maximum is located.  $E$  corresponds to the activation energy of the glow peak, directly related to the width of the full-width half maximum. The arrow below the activation energy is intended to indicate that this parameter determines the width of the glow peak in the first-order kinetics model.

**Batch file handling.** The GCA software begins by requesting user input of a directory containing the CSV input files. Using the C++17 Filesystem library, the software accesses the number of CSV files present in the root directory recursively and prompts the user to confirm its findings. The files are then opened one at a time, with raw data saved into the corresponding vectors. The time complexity of the batch handling process implemented for this code is determined by the number of files,  $n$ , and the number of lines in those files,  $m$ , for a time complexity of  $O(n^m)$ .

**Data noise reduction.** A preliminary analysis indicated that data containing large amounts of noise or sudden spikes in the signal caused convergence failure. As a result, a number of smoothing methods were explored and tested. The moving average smoothing algorithm was chosen to smooth the data (Klopfenstein 1998). This average smoothing algorithm averages values based on a variable number of neighbors. In this implementation, the five nearest neighbors are used. One iteration of the average smoothing algorithm has a time complexity of  $O(n)$ . A moving average algorithm is then run iteratively five times, making the complexity  $O(n^5)$ .

**Automated peak detection.** The most unique feature of this GCA software is the automatic identification of glow peaks and their associated fitting parameters. Historically, this has been the step in GCA software that requires the most human attention, requiring the researcher to analyze individual spectra one at a time. A researcher would estimate the temperature at each glow peak maximum and intensity and then approximate the activation energy for each of the glow peaks. The presented GCA software employs an automated method for determining glow peak locations and fitting parameter estimates.

**Peak detection through subtraction.** The first attempt at identifying glow peaks was based purely on the identification of global maxima and subtraction of the globally largest peaks from the signal. Preliminary results indicated that this was not effective. Often, subtracted peaks were found to be in the wrong location, resulting in too many or too few identified peaks.

**Peak detection through derivatives and subtraction.** Peak identification begins instead by finding all local maxima, minima, and inflection points. These features are identified using first and second derivatives of the input. Derivatives are created in  $O(n)$  time using the one-dimensional five-point stencil method (Mai-Duy and Tran-Cong 2013). The local maxima and inflection points are then subject to an iterative filtration process, eliminating any maxima or inflection points resulting from noise. Next, all peaks remaining are passed to the FOK model and subtracted from the original signal. If the integrated counts after this subtraction are above a

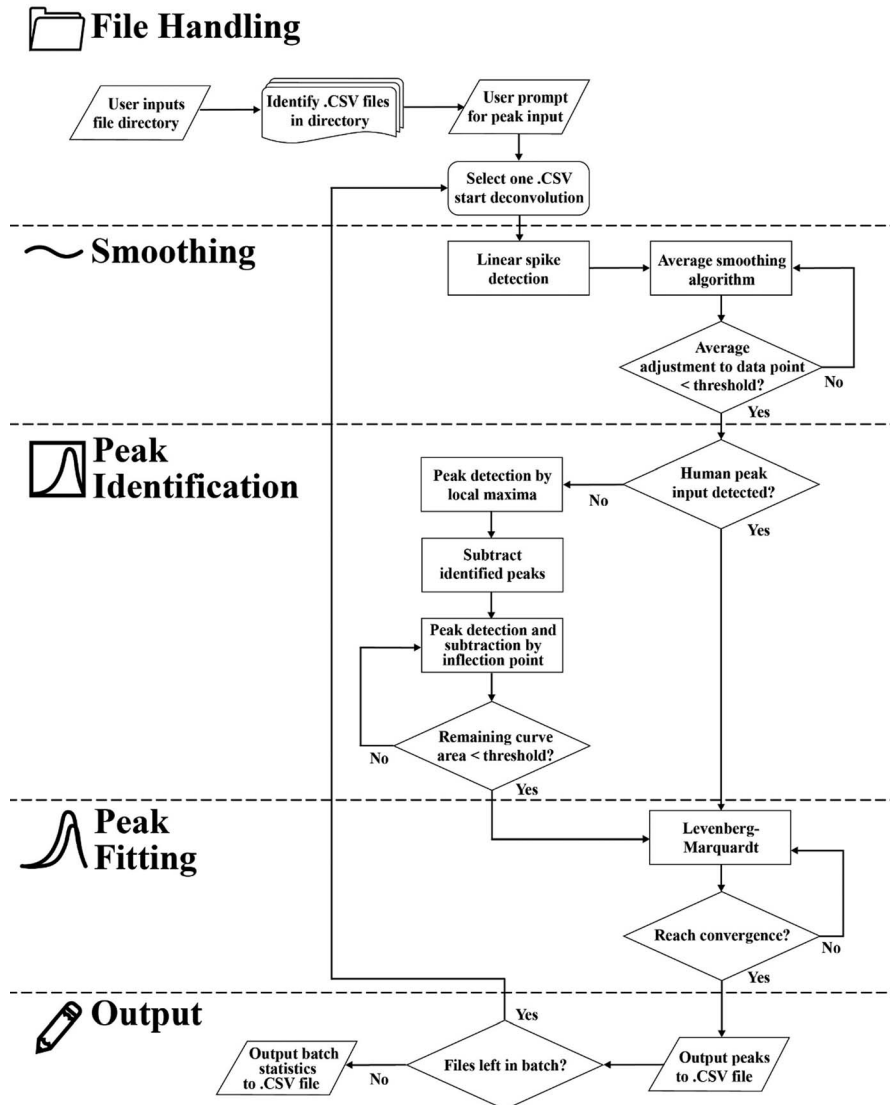


Fig. 2. Flow chart detailing the process flow of the glow curve analysis software.

threshold, the maxima, minima, and inflection points of the remaining signal are again identified and vetted. Each successively identified peak is passed to the FOK model and subtracted from the remaining signal. This process is iteratively repeated until the remaining signal is reduced below a threshold. Finally, the initial guesses for peak parameters are then passed into the LMA.

**Peak fitting using Levenberg-Marquardt algorithm (LMA).** The peak identification method described above generates approximate peak parameters, similar to human input. For analysis, these initial parameters must be refined, presenting a non-linear least squares minimization problem. LMA is a common solution to such problems and relies heavily on matrix mathematics (Moré 2006; Bellavia et al. 2018). The software uses a modified iterative LMA, which

minimizes the error between the original emission spectra and the summation of the fitted glow peaks. LMA is run iteratively until convergence is met or failure occurs due to iterative exhaustion.

**Figure-of-merit.** The chief metric used to assess the goodness of a glow curve fitting is called the figure-of-merit (FOM) (Balian and Eddy 1977; Bos et al. 1993). This FOM is the average of the percent difference between the fitted points and measurements, as given by:

$$FOM = \sum_{j_{start}}^{j_{stop}} \frac{|y_j - y(x_j)_j|}{A} \times 100, \quad (3)$$

where  $j_{start}$  is the initial temperature in the fit region,  $j_{stop}$  is the ending temperature in the fit region,  $y_j$  is the photomultiplier tube (PMT) current at temperature  $j$ ,  $y(x_j)$  is the value of the fit function at temperature  $j$ , and  $A$  is

the area under the peak for glow curves. The original publication on this FOM for GCA describes a “good fit” to have a FOM less than 3% (Balian and Eddy 1977), while others describe a “few percent” as a good fit (Horowitz and Yossian 1995).

**Output.** Upon completion of the LMA algorithm, a list of identified peaks and their respective curve areas are created. The separated peaks are written to an output file in CSV format and placed in an automatically created batch output folder. The output file can be easily plotted. A batch statistics file is also created, containing a list of information pertaining to each input file, including sample barcode, heating rate, FOM, total curve area, and a list of the areas under each glow peak. This batch file is intended to allow for the analysis of trends among samples.

## Materials

**Thermoluminescent dosimeters.** The thermoluminescent dosimeters (TLDs) used in this paper were LiF:Mg,Ti chips (TLD-100; ThermoFisher Scientific, Waltham, MA). These chip-type TLDs have dimensions of 3.2 mm wide by 3.2 mm long by 0.89 mm high. A single set of 100 TLDs was used to create all data in the time-temperature profile and dose variation experiments. A number of historical glow curves from LiF:Mg,Ti and CaF<sub>2</sub>:Dy (TLD-200) dosimeters were used for the time complexity experiment described below.

**Thermoluminescent dosimeter reader.** The REXON UL-320-FDR thermoluminescent reader (UL-320-FDR; REXON, Beachwood, OH) was used to obtain the glow curves analyzed in this paper. This reader automates the TLD readout process through the use of contact heated planchets with an infrared temperature feedback system (Kuchta et al. 2020). This TLD reader integrates photon counts over time intervals of 0.1 s during heating for all measurements used in this paper.

**Irradiation process.** An ~269 GBq <sup>137</sup>Cs irradiator (Model 28-8A Irradiator; J.L. Shepherd and Associates, San Fernando, CA) that had been fully characterized (Boria et al. 2017; Mapes et al. 2018) and undergone thorough quality control (Noey et al. 2021) was used for the experiment. Doses ranging between 2.4 mGy and 30 mGy were delivered, corresponding to irradiation times of 5.5 min to 68 min, respectively. Dosimeters were irradiated on a standard 40-cm × 40-cm × 15-cm-thick polymethyl methacrylate (PMMA) irradiation phantom (Parker et al. 2011) located at 1 m from the source.

**Personal computer.** All results reported in this paper were generated using a stock 2015 Macbook Pro (A1398, Apple Inc, Cupertino, CA) running macOS Catalina version 10.15.7 using the Terminal application with clang version

12.0.0. All runtime measurements were taken using the on-board real-time clock. The code was also tested and performed equivalently on Windows and Linux systems.

**GlowFit.** A freely downloadable first-order kinetics GCA software based on Levenberg-Marquardt minimization was used to provide a point of comparison for the presented GCA software’s performance. The software used for comparison is called GlowFit (Puchalska and Bilski 2006). It was chosen for its pleasant graphical interface and ease of download.

## Experimental methods

**Experimental motivation.** For any piece of software, it is critical to assess its performance under real conditions. Two factors that cause variance in TLD output signals are the dose applied and the heating rate used at readout. If a lower dose is applied, the glow peaks have a lower intensity. Conversely, if a higher dose is delivered, the peaks will have a higher intensity. Furthermore, if a higher heating rate is used, the glow peaks will appear to be closer together because TLD readers have a fixed sampling rate. The opposite is true of lower heating rates. Thus, in order to test the GCA program on the most diverse set of inputs, two experimental datasets were created to test both of these factors.

**Time-temperature profile variation experiment.** The first dataset was generated to test the GCA program’s performance on a variety of heating rates. The set of 100 LiF:Mg,Ti chips were annealed at 400°C for 1 h in a benchtop muffle furnace (Thermolyne Type 1300; ThermoFisher Scientific, Waltham, MA). This set of 100 TLDs was then irradiated on the phantom to an air kerma of approximately 15 mGy. The 100 TLDs were grouped into 10 sets of 10 TLDs, and each set was then read out at differing time-temperature profiles (TTPs). This process was repeated three times, representing 30 sets of 10 files. However, only 22 sets of 10 were used for this analysis, representing 220 files. The TTPs used in this experiment began with a 5 s increase to 40°C. Next, a linear heating rate between 1°C s<sup>-1</sup> and 6°C s<sup>-1</sup> increased the temperature to 250°C. All TTPs ended with a 5 s hold at 250°C. Due to the variable readout length, the minimum time between irradiation and readout was 10 min, and the maximum time was 24 h. A mild correction was applied to account for this. The reader settings during the experiment were held constant throughout. The photomultiplier tube high voltage was 1,200 V with a max dark current of 50 counts and a maximum light current of 1,000,000 counts. Thermoelectric contact heating was employed, with a recommended continuous flow of N<sub>2</sub> gas with a pressure of 20 psi used to decrease chemoluminescence and maintain a low dark current for all measurements (Kuchta et al. 2020). The resulting dataset of TLD signals was then processed using

the GCA software running on the MacBook Pro previously described. This dataset was also fed to GlowFit for contrast.

**Dose variation experiment.** The second dataset was generated to test the performance dependence on dose. The same 100 TLDs were oven annealed as described above. TLDs were then irradiated on the phantom in groups of 10 for between 5.5 min and 68 min. These times correspond to air kerma values between 2.4 mGy and 30 mGy. The time between irradiation and readout was between 30 min and 4 h. All readouts were conducted under the same TTP for this experiment. This TTP begins with a 5-s preheat to 40°C, then a 155-s linear rise to 250°C, and finally, a 5-s hold at 250°C. The reader settings described in the previous section were used for this experiment also. As in the first experiment, these files were used as a testing dataset for the GCA program and also processed using GlowFit for comparison.

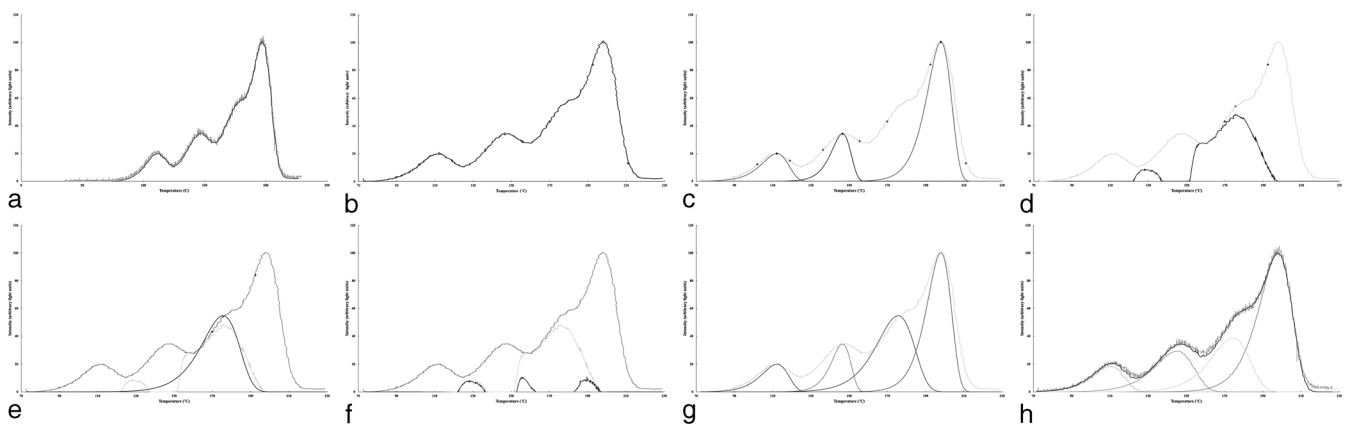
**Time complexity motivation.** During preliminary testing, it was discovered that the runtime of the GCA software has a strong dependence on three factors: the number of input files, the length of the input files, and the noisiness of the input files. Therefore, a dataset using a large number of files to test the relation between file length and runtime was created. The noisiness of a specific file was not measured or calculated directly; however, files showing prohibitively long runtimes were visually examined and removed from the dataset.

**Time complexity experiment.** A total of 2,377 input files were used for the purpose of testing the average runtime and time complexity of the GCA program. These spectra were procured from the same TLD reader used in the previous two experiments. These data come from a large

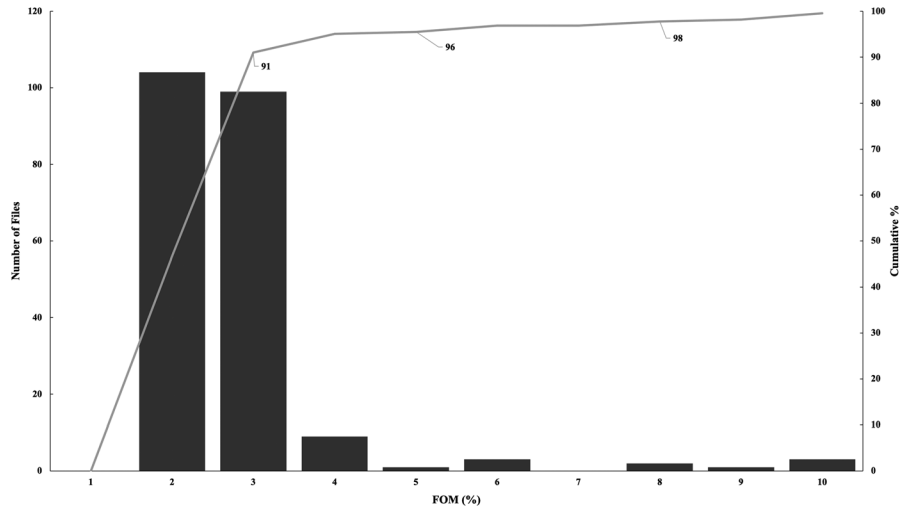
number of experiments using a variety of reader settings and TLDs. TLD-100 glow curves constitute 87% of this dataset, with the remainder consisting of TLD-200. In this dataset, TLDs received a variable amount of air kerma and were read out with a diverse assortment of TTPs. This dataset was run only with the GCA program. First, the GCA software was fed a variable number of files from the large dataset in random order. Total runtimes were measured to determine any relationship between the number of files input and the total runtime. Second, the entire dataset was run, each file runtime was measured, and the number of rows in the file counted.

## RESULTS

The total size of the GCA program accompanying this paper is 339 kB, which is incredibly lightweight. Fig. 3 outlines the intermediate steps of the glow curve analysis software. First, the initial signal is read into the software. Next, the data are smoothed, as depicted in Fig. 3a. In Fig. 3b–f, an example peak identification process is portrayed. These figures include identification of local maxima and inflection points, the initial identification of only three glow peaks, and the signals remaining after subtraction of those identified peaks. Fig. 3g shows the final output of the peak identification process with all four commonly identified TLD-100 glow peaks correctly found (Harvey et al. 2011; Horowitz and Yossian 1995). Fig. 3h illustrates the GCA output following LMA with well-fit glow peak output. The TL signal is closely approximated by the summation of the separated peaks with a FOM of 2.25%.



**Fig. 3.** The glow curve analysis program process flow, showing (a) initial operation of the average smoothing algorithm with the original input signal (light gray) and the input signal after smoothing (black), (b) the identified local maxima (light grey circles) and inflection points after vetting (dark grey diamonds), (c) the first three identified glow peaks (black and dark grey) after initial fitting parameters have been generated, (d) the difference in signal (black) between the first three identified glow peaks and the original signal with the nearby inflection points and maxima (grey), (e) the fourth identified glow peak (black), (f) the remaining signal difference (black) after the subtraction of the fourth peak (this signal is found to be below a set threshold; thus the peak identification algorithm terminates), (g) four peaks recognized by the peak identification algorithm (dark grey; these peaks are now passed into Levenberg-Marquardt algorithm for iterative fitting), (h) the final program output depicting the original input signal (light grey), the fitted glow peaks, (grey) and their sum (black). The final fitting has a figure-of-merit of 2.25%.

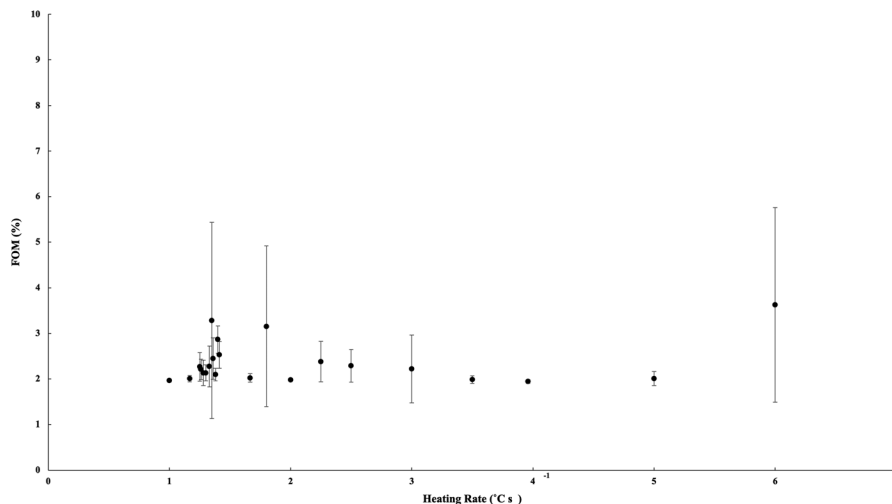


**Fig. 4.** Fractional figure-of-merit among the 220 files used in the time-temperature profile variation experiment. The dark grey bars behave as bins with a thickness of 1, containing FOM values within 0.5 of the number shown. Values are placed in the upper bin if the value is 0.4999 greater than the bin number. One file having a FOM of 29.8% was omitted from this figure, it was determined to be a noisy outlier. Shown in light grey is the cumulative percentage of files having FOM below selected values. Notably, 91% of the data has a FOM of 3.5% or lower.

#### Time-temperature profile (TTP) variation experiment

The glow curve analysis of the acquired signals using the experimental method above was completed in less than an hour with a 100% convergence rate. The peak identification algorithm successfully located the four observed glow peaks of TLD-100 for 97.7% of the input spectra. There were five spectra for which the GCA program did not find four peaks. Three spectra separated into five glow peaks, and two spectra finished with three peaks. A minor modification of the peak identification algorithm might resolve this 2.3% misidentification rate. The average FOM was 2.6%. The FOMs ranged from 1.9% to 29.8%. This final value is an outlier stemming from excessively noisy input data. The next highest FOM is 9.8%, as shown in Fig. 4, which details the distribution of FOM for the whole dataset

without the highest outlier. It is notable that 91% of the dataset has a FOM of 3.5% or less. The average FOM of each group of 10 TLDs is plotted as a function of the linear heating rate in Fig. 5. This result demonstrates that there is no relationship between FOM and heating rate for the GCA program. GlowFit also processed this dataset and produced an average FOM of 2.9% based on user-created initial guesses and ranges. GlowFit failed to produce any output for the noisy outlier commented on previously. This result is extremely close to the 2.6% average found by the GCA software. This demonstrates that the GCA program presented is comparable to other previously published GCA programs of the same type. The low value of the average FOM and high peak identification rate indicate that the GCA software is very successful for TLD-100 at a dose of



**Fig. 5.** Fractional FOM as a function of heating rate, demonstrating that the heating rate does not affect the glow curve analysis software's performance and the TLD reader's consistency.

15 mGy, irrespective of the readout process. This experiment effectively demonstrates the robustness of the GCA software for variations in TLD-100 glow peak locations.

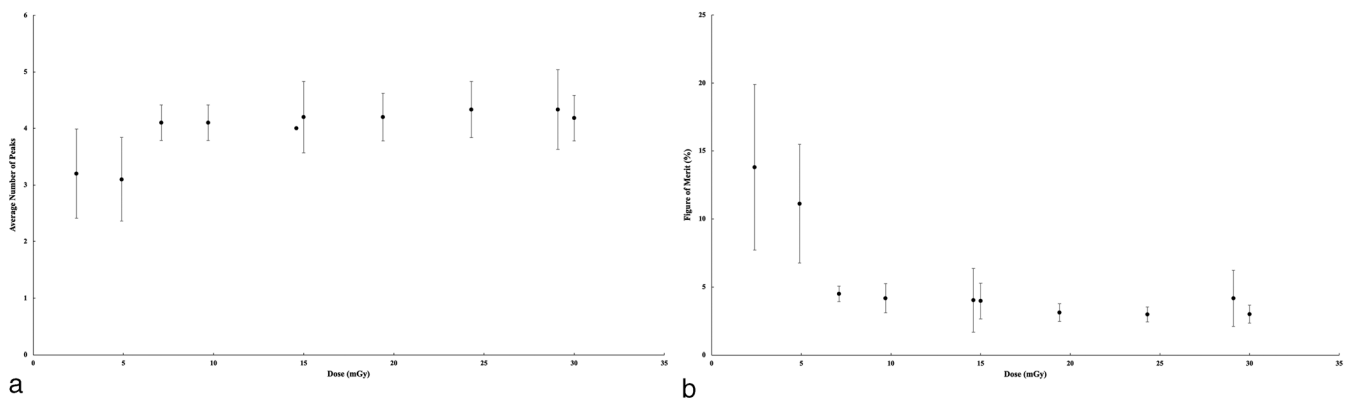
### Dose variation experiment

The glow curve analysis of these 100 TLD spectra was completed in under an hour with a 98% convergence rate. The two files for which it did not converge contained signals below a set threshold for peak identification. The peak identification algorithm correctly located four glow peaks in 68% of the spectra. It incorrectly found that 4% had two peaks, 12% had three peaks, and 15% had five peaks. There is a correlation between low doses and the number of peaks identified, as shown in Fig. 6a, indicating that the peak identification algorithm should be modified to better assess air kermas lower than 7 mGy. Above 7 mGy, there is no clear correlation between dose and the number of peaks identified. The average figure-of-merit was 5.5%, with a minimum FOM of 1.93% and a maximum of 25.21% (again a noisy outlier). As dose varied from 2.4 mGy to 30 mGy, the relationship between FOM and dose was graphed in Fig. 6b. It is clear from this plot that below 7 mGy, there is a correlation between FOM and dose. There may be a weak relation between higher dose and lower FOM; however, such a statement is outside of the uncertainty interval of this experiment. GlowFit produced an average FOM of 7.5% for this dataset. Again, the presented average FOM of 5.5% compares well to this number. Based on these results, the GCA performs as intended above 7 mGy. However, it is not recommended that this GCA program be used for TLD-100 at doses below 7 mGy without human input. For doses above 7 mGy, this experiment definitively shows the reliability of the GCA software for variations in TLD-100 glow peak height.

### Time complexity experiment

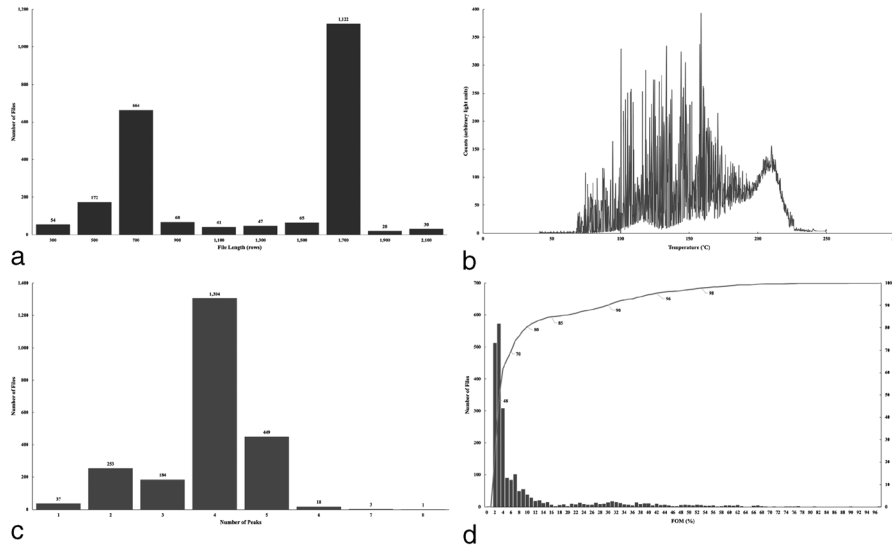
The overall runtime of this program depends strongly on the number of input files and input file length. For this work, 2,377 CSV input files having an average length of 1,168 rows were processed. The distribution of input file length is shown in Fig. 7a. The convergence rate was approximately 95%. The files that did not converge were overly noisy and had a runtime that was prohibitively long. For example, the Levenberg-Marquardt algorithm took more than 39 min to run one iteration successfully on the signal plotted in Fig. 7b. When visually examined, it is clear that the data in these files is primarily resultant from non-TLD phenomena. For this reason, this signal and others like it were aborted during glow curve analysis. The average number of peaks identified for this dataset was 3.87. Fig. 7c shows the distribution of the number of files that presented between 1 and 8 glow peaks. Analysis of this distribution indicates that the average number of glow peaks identified in the TLD-100 files was 4.1 peaks. Approximately 63% of the TLD-100 input files successfully found 4 glow peaks. For the TLD-200 spectra, the average number of identified peaks was 2.5. This clearly indicates that the peak identification algorithm is not well calibrated for TLD-200. However, a large number of these spectra are the result of aberrant readout conditions. As a result, this does not measure the effectiveness of the peak identification process properly. The average FOM of this dataset was found to be 9.4%. A histogram of FOM is shown in Fig. 7d. This distribution is multimodal, resulting from the heterogeneity of the input. These results are presented only for reference; they are not suggested to demonstrate the performance of the GCA software due to the abnormality of the dataset.

The total runtime of the software is directly linear with respect to the number of files input. This result is shown in Fig. 8a. The average length of a file in this dataset is 1,168



**Fig. 6.** Results of the dose variation experiment. Shown are (a) the average number of discovered peaks as a function of the dose, and (b) the relationship between FOM and dose. It is notable that there is a dramatic drop in the number of peaks identified below 7 mGy. Above 7 mGy, there is little to no change in the number of peaks identified. As a result, the files below 7 mGy have higher FOM owing to the lower number of peaks identified, and at higher doses, the change in FOM is lower than the uncertainty.





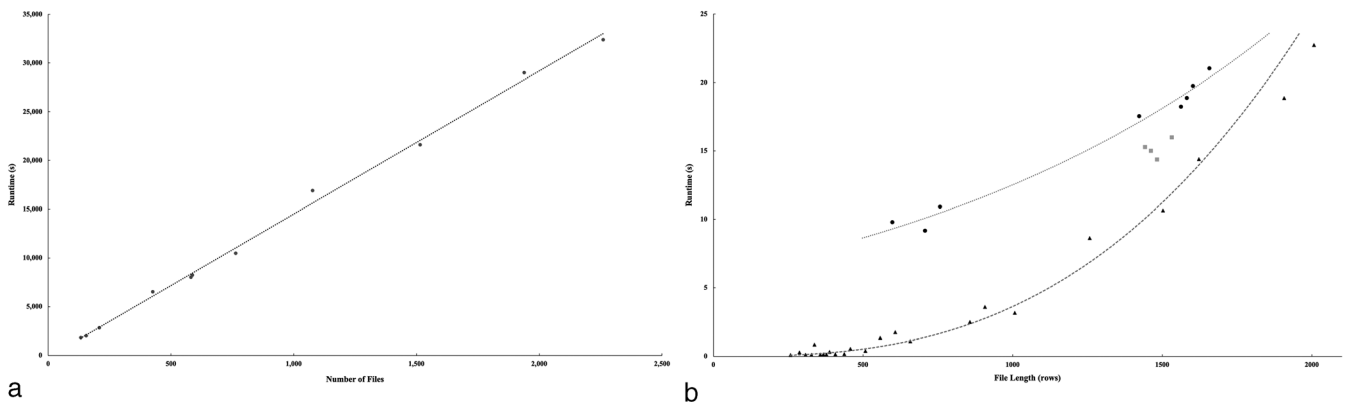
**Fig. 7.** Statistics regarding the challenging nature of the time complexity dataset, including (a) the distribution of the lengths of the input files in terms of rows (the bars in this plot behave as bins with a thickness of 200 centered around the number given), (b) an example of a highly aberrant signal that caused exorbitantly long convergence time when passing through the Levenberg-Marquardt algorithm, (c) the quantity of files for which a certain number of peaks was identified (because the number of identified peaks are integers, there is no width to these bins), and (d) the distribution of FOM over this large heterogeneous dataset (dark grey) with the cumulative percent of files at each bin (light grey). It is noted that such a dataset would be inappropriately challenging for algorithm performance testing.

rows, and the corresponding average runtime was fit to be approximately 14.7 s with a standard error of 0.046 s. The Levenberg-Marquardt algorithm dominates file runtime. This is clearly demonstrated when the individual file runtimes are recorded. Runtimes of files with the same length were averaged to create Fig. 8b. The function describing the increase in runtime is exponential with respect to the number of rows. As previously mentioned, the GCA software processed TLD-100 and TLD-200 spectra differently. The principal difference pertains to the number of identified and therefore fit peaks. On average, TLD-200 spectra had 1.6 fewer identified peaks. Therefore, TLD-200 spectra are, on average, processed faster than TLD-100 spectra due to

fewer Levenberg-Marquardt steps. This is an important result, indicating that the runtime of any file is strongly linked to the number of rows input as well as the number of glow peaks being fit.

## CONCLUSION

The GCA software tool described in this paper is the first step toward fully-automated glow curve analysis. The software is incredibly lightweight and is compatible with all major operating systems running C++17. The GCA tool is fast, with an average runtime of approximately 15 s for a 1,200-row input file. The runtime of the program depends



**Fig. 8.** The runtime of the program (a) as a function of the number of input files, demonstrating a linear increase, and (b) averaged for files of the same length with trendlines added to guide the eye. The upper curve shows the exponential increase in runtime for LiF:Mg,Ti spectra versus the lower curve displaying an exponential increase in runtime for CaF<sub>2</sub>:Dy spectra; the black circular points are primarily resultant from LiF:Mg,Ti spectra, the black triangles primarily are resultant from CaF<sub>2</sub>:Dy spectra, and the grey squares are a mix of these spectra.

linearly on the number of input files. The runtime of a specific file depends on the number of rows exponentially. Runtimes also depend on the number of glow peaks being fit per file and the noisiness of the input files. The output quality is comparable to those produced by other available GCA software. The GCA program has a 98% correct peak identification rate for TLD-100 at 15 mGy, independent of the readout heating rate. This software is not recommended for use at doses below 7 mGy without human input. This code presents a framework for future research and improvement in fully-automated peak identification.

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## REFERENCES

- AQ5 Azorín NJ. Thermoluminescence dosimetry (TLD) and its application in medical physics. *AIP Conference Proc* 724:20; 2004.
- Balian HG, Eddy NW. Figure-of-merit (FOM), an improved criterion over the normalized chi-squared test for assessing goodness-of-fit of gamma-ray spectra peaks. *Nucl Instrum Methods* 145: 389–395; 1977.
- Basun S, Imbusch GF, Jia DD, Yen WM. The analysis of thermoluminescence glow curves. *J Lumin* 104:283–294; 2003.
- Bellavia S, Gratton S, Riccietti E. A Levenberg–Marquardt method for large nonlinear least-squares problems with dynamic accuracy in functions and gradients. *Numerische Mathematik* 140: 791–825; 2018.
- Boria AJ, Rucinski BD, Abraham SA, Dawson AS, Jawad AH, Miklos JA, Kearfott KJ. Cs-137 Dosimeter irradiation facilities: calibration frequency, precision, and accuracy. *Health Phys* 112:357–363; 2017.
- Bos AJJ, PETERS TM, Gomez Ros JM, Delgado A. An intercomparison of glow curve analysis computer programs. I. Synthetic glow curves. *Radiat Protect Dosim* 47:483–487; 1993.
- Chen R. On the calculation of activation energies and frequency factors from glow curves. *J Appl Phys* 40:570–585; 1969.
- AQ6 Cobham A. The intrinsic computational difficulty of functions. Logic, methodology and philosophy of science: proceedings of the 1964 International Congress (studies in logic and the foundations of mathematics). North-Holland Publishing 24–30; 1965 .
- Harvey JA, Rodrigues ML, Kearfott KJ. A computerized glow curve analysis (GCA) method for WinREMS thermoluminescent dosimeter data using MATLAB. *Appl Radiat Isot* 69: 1282–1286; 2011.
- Horowitz YS, Moscovitch M. Computerized glow curve deconvolution applied to high dose ( $10^2$ – $10^5$  Gy) TL dosimetry. *Nucl Instr Meth Phys Res* 243:07–214; 1986.
- Horowitz YS, Yossian D. Computerised glow curve deconvolution: application to thermoluminescence dosimetry. *Radiat Protect Dosim* 60:1, 3; 1995. AQ7
- International Organization for Standardization. IEC 14882:2017 Information technology—programming languages—C++. pub-ISO 5:1605; 2017. AQ8
- Kitis G, Gomez-Ros JM, Tuyn JWN. Thermoluminescence glow-curve deconvolution functions for first, second and general orders of kinetics. *J Phys D Appl Phys* 31:2636–2641. 1998.
- Klopfenstein R. Data smoothing using a least squares fit C++ class. *ISA Trans* 371:3–19; 1998.
- Kuchta JR, Thiesen JH, Noey JD, Chung Long Kiu, Kearfott KJ. Preliminary experiences with the Rexon UL-320-FDR: an automated thermoluminescent dosimeter reader with removable contact heating planchets and an infrared temperature feedback system. *Health Phys* 2020 (in press). AQ9
- Lilley E, McKeever SWS. On the order of kinetics for thermoluminescence in LiF (TLD-100). *J Appl Phys* 16(2); 2000.
- Mai-Duy N, Tran-Cong T. A compact five-point stencil based on integrated RBFs for 2D second-order differential problems. *J Computational Phys* 235:302–321; 2013. AQ10
- Mapes JL, Liu K, Abraham SA, Wilhelm AS, Latosz LV, Kearfott KJ. Setup and characterization of a cesium-137 dosimetry calibration source in a space-constrained environment. *Health Phys* 115:569–580; 2018.
- Moré JJ. The Levenberg-Marquardt algorithm: implementation and theory. In: *Numerical analysis*. Berlin, Heidelberg: Springer; 1978:105–116. 2006. AQ11
- Moscovitch M, Horowitz YS, Oduko J. LiF thermoluminescence dosimetry via computerised first order kinetics glow curve analysis. *Radiat Protect Dosim* 6.1-4:157–159; 1983. AQ12
- Noey JD, Golduber RM, Kearfott KJ. Analysis of long-term quality control data for a  $^{137}\text{Cs}$  dosimetry calibration Source. *Health Phys* 120:227–242; 2021.
- Parker LW, Harvey JA, Kearfott KJ. An integrated system for the beta, gamma and neutron calibration and storage of thermoluminescent dosimeters for a research laboratory. *Health Phys* 100(Suppl 1):S43–S49; 2011.
- Puchalska M, Bilski P. GlowFit—a new tool for thermoluminescence glow-curve deconvolution. *Radiat Meas* 41:659–664; 2006. ■ ■

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