**A Computerized Glow Curve Analysis Software written in C++**

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**Abstract**

The examination of thermoluminescent dosimeter glow curves is useful for the detection of abnormalities in dosimeter processing. When appropriately analyzed, glow curves hold added information about thermoluminescent material behavior. The mathematical separation of a glow curve into contributions from energetically different trap states, or glow curve analysis (GCA), may be utilized to remove undesired effects of signal fading for complex materials. GCA illuminates the material-specific models of thermoluminescence. In special circumstances, it could lead to improved statistical performance at low doses. Generalized computerized GCA software for the deconvolution of glow curves was written in C++. The code optimizes the fitting process utilizing either a user-specified first-order kinetics model or a one trap-one recombination level model. The code was tested using experimental glow curve data from LiF:Mg,Ti, CaF2:Dy, CaF2:Tm, and CaF2:Mn. Glow curves from a variety of different readers may be analyzed with the code. Output consists of a list of fit parameters and deconvoluted gaussian data in comma-separated-value form for easy graphing and analysis. The adaptation of the code for data obtained using complex time temperature profiles may be possible.

**Keywords**: dosimetry, first order kinetics, glow curve, Levenberg Marquardt

**1. Introduction**

Radiation dosimetry is a fundamental field in Medical Physics, specifically the application of radiation and radioisotopes. Ionizing radiations most used in

diagnosis and radiotherapy are X-rays, gamma radiation and beta particles; dosimetry attempts to measure the risk and effectiveness of these radiation exposures [(Azorín,2004)](#References). The most appropriate data collection methods in dosimetry pertain to the study of thermoluminescent dosimeters (TLD), which utilize the thermoluminescent (TL) phenomenon, which describes particular irritated solids which emit light when heated at a temperature below their incandescence temperatures [(Reuven, 1997)](#References).

A critical tool utilized for the study of TLDs is glow curve analysis, the deconvolution of individual glow curves from the emitted thermoluminescent spectra into individual glow peaks. Figure 1 depicts an example deconvolution from our GCA software, the outer pink curve being the original glow curve, and the four individual glow peaks representing the energy released from a trap state when heated to a particular temperature. Traditional methods of deconvolution involve the manual input of initial fitting parameters, which are susceptible to convergence failures. Turning the process of yielding convergence and accurate glow curves into more of an art form than a science. Our work consists of developing a more intelligent Glow Curve Analysis (GCA) software which automates the process of identifying and fitting the individual glow peaks of the TL spectrum to eliminate human participation.

The fitting metric most widely accepted when discussing glow curve analysis is the Figure of Merit (FOM), a measurement of the percent error in the final fit against the original emission spectra [(Bos et al., 1994)](#References).

*Equation 1 – FOM*

where *jstart* is initial temperature in the fit region, *jstop* is the ending temperature in the fit region, is the photomultiplier tube (PMT) current at temperature j, is the value of the fit function at temperature j, and A is the area under the peak. The original publication of the FOM describe a “good fit” to have a FOM less than 3% [(Balian and Eddy, 1977)](#References), and any value on the order of a few percent to be an accurate fit produced by a GCA software [(Horowitz and Yossian, 1995)](#References).

Thus far the automated GCA software has been able to yield superior FOM’s, convergence rates, run times, and batch file handling capabilities then other popular fitting software’s requiring human input initial fitting parameters. Performance testing averages four minutes to process a batch directory of one hundred LiF:Mg,Ti (TLD-100) emission spectra yielding sub 3% FOMs, with a mean FOM of 1.58%, well below the industry standard. A mean convergence rate of 98%, and average 2% of analysis which failed to convergence still on yielding sub 10% FOMs. Common causes for failure to converge will be discussed further in our findings.

**2. Methods**

**2.1. Software Specifications**

The GCA software is written entirely in C/C++11, the decision to utilize the C++ language verses other competitive languages was to provide superior processing speeds, cross platform compatibility, and a very lightweight distribution. Utilizing purely the Standard Template Library, which comes stock equipped on all modern computers. With one exceptions being the use of the C++ Boost Filesystem Library for cross platform batch file handling, however the software comes equipped with the compiled binary file for the particular function utilized. Allowing users who follow the proper single step download procedures, as describe in the Read Me file, to seamlessly download and run the software on any Windows, Linux/Unix, or any other C++11 equipped system.

The software is currently limited to only processing files which are formatted as comma-separated values(CSV). While this may come to some users as an inconvenience; in order to maintain performance, low level optimizations were made specific to CSV file processing. The most common modern dosimeters in use provide the option for read outs to be CSV formatted; for grandfathering in previously ill formatted spectra readouts, file conversion software is easily accessible through Excel and other online resources.

The software processes files through a five-stage system consisting of: batch file handling, data noise reduction, automated peak detection, curve fitting, and output. The five-stage system can be visualized by the flowchart in figure 2. Each stage of the process has undergone extensive planning, implementation, and revision with the goal to improve FOMs, while maintaining above average processing speeds relative to other popular manual input software.

**2.2. Batch File Handle**

One of the most superior features of the software is its ability to process hundreds of emitted spectra rapidly. Utilizing the automated peak detection, the software does not require any user input other than the input directory housing the properly formatted spectra files. The batch file system uses an advanced CSV reading and writing method taking advantage of optimized low-level tools built into the more primitive back compatible C language. Paired with the C++ Boost Filesystem library the software accesses the number of CSV files present in the provided directory, prompts the user to confirm its findings in the directory and to begin deconvolution.

Batch handling runs at very optimized efficiency, as discussed sacrificing multiple input file formats for speed. The complexity of this stage of the system runs in technically, using big-O notation, in . However due to the nature of this stage the complexity relies on the number of files, *n,* and the number of lines in those files, *m,* making the complexity . Runtimes for this stage were collected by running the software with a system clock variable from the start of the code to the beginning of the deconvolution stages, and then again after the deconvolution during the output stages. Processing one hundred files required 4.2 seconds of read-in time, and 6.1 seconds of write-out time.

**2.3. Data Noise Reduction**

The main, and on average the only, problem that causes the software to fail to converge is overly noisy data read in from the dosimeter. Sudden spikes during sampling can be cause by a wide variety of external sources, ranging from light exposure to dust particles in the air. General minor noise can result from dosimeter calibration, dosimeter age, and dosing techniques. These phenomena can prove very problematic for the later peak identification and fitting stages, and so a wide variety of smoothing methods had to be explored and tested. Resulting in a multi-stage smoothing system which has been able to get convergence rates of near 100%.

The software prior to employing mathematical smoothing techniques does a , linear, spike detection stage. A single pass through the data removes any dramatic spikes in data surpassing a variable threshold based on averaging sections of data. This step is employed to prevent unnecessary iterations in the next iterative mathematical smoothing process, reducing overall complexity and dramatically reducing runtimes.

Proceeding the spike detection stage the software employs a moving Average Smoothing algorithm [(Klopfenstein, 1998)](#References) to filter out smaller point to point noise. A wide variety of smoothing algorithms were tried and tested including but not limited to: Bin Smoothing, Running line (local and global), Kernal Density ([Bin and Matt, 1994](#References)), and LOESS ([Simonoff, 2012](#References)). While some methods produced similar, and in some cases even slightly improved, smoothing results the Average Smoothing algorithm yielded consistent rapid results that felt most appropriate to the context. This work will only spend time discussing the Average Smoothing algorithm at length. Described by [(Klopfenstein, 1998)](#References) as,

*where*:

|  |  |
| --- | --- |
| *i,* | *index into sliding window* |
| *m,* | *sliding window width* |
| *n,* | *number of raw data points* |
| *s,* | *smoothed data array* |
| *x,* | *raw data entry* |
| *j,* | *index into raw data* |
| *note:* | *j ranges from (m-l)/2 to n-(m-1)/2* |

*Equation 2 – Average Smoothing algorithm*

The Average Smoothing algorithm is a simple to implement and lightweight smoothing algorithm which averages a value based on a variable number of neighbors. The software determines the number of data points and assess the appropriate number of neighbors that can be averaged from. Making one iteration run at a complexity of . The algorithm is then run iteratively, *m* times, until the alteration to a data point falls below a reasonable threshold, or hits a maximum iterations, resulting in a complexity of .

**2.4. Automated Peak Detection**

What ultimately sets the GCA software apart from its competitors is the previously discussed ability for the software to automatically and intelligently identify the target peaks and determine their fitting parameters for the fitting stage. Historically this has been the step in manual GCA software which becomes the bottleneck. The process for manually determining fitting parameters requires the researcher to: analyzing individual spectra one at a time, guess the temperature at the not yet deconvolved glow peak’s maximums, estimate the maximum intensity for each glow peak, and then calculate a activation energy for each of the glow peaks based on these estimates. This method when conducted by hand can prove difficult, and in some case impossible, to yield convergence.

The automated GCA software employs a mathematical method for determining where the glow peaks are located, and determining highly accurate fitting parameter estimates. Additionally, the software determines an error margin on each estimate which will be utilized during the fitting stage to cut down fitting iterations, and more frequently yield convergence.

The process for peak detection creates both an array of the first derivatives at each data point along with an array of second derivatives. The derivatives are accurately calculated using first and second order a one dimensional five-point stencil ([Mai-Duy and Tran-Cong, 2013](#References)).

*Equation 3 – First Order Five-Point Stencil*

*Equation 4 – Second Order Five-Point Stencil*

Where, *h,* is a step size, however due to the nature of our use *h* was always equal to one.Then using these arrays a list of local maximums and inflection points are determined, and sent through a iterative vetting process. The vetting process attempts to eliminate false maximums and inflection points which arise as a result of any remaining noise in the data. Figure 3 depicts the glow peak maxima and inflection points the software manually determined after vetting the lists of local maxima and inflection points.

Once a list of maximums and inflection points has been established the first stage of a two part pipeline for determining fitting parameter begins by making an initial guess at a peak fitting based on each of the maximums. The maximums, and now their respective fitting parameters, are then checked further to ensure they did not result due to any noise left in the data. The remaining maximums and their fitting parameters are passed to a temporary fitting model to produce rough estimate glow peaks, fitting models will be further discussed later in our findings. Each of the temporary peaks are then subtracted away from the original glow curve, and of any remaining area under the glow curve is then used to determine peaks between local maxima.

For determine peaks remaining after the subtraction of the peaks located at local maxima a similar method as described using the list of maximums is used for the list of inflection points. The nearest inflection point to the maximum remaining data point is fit using the same fitting methods as if it was another local maximum; the resulting glow peak is then additionally subtracted away from the original glow curve. Figure 4 shows the resulting glow curves after each glow peak is subtracted away from the original output spectra; the maximum value of the remaining curve, found on the blue curve, is then used to identify the nearest inflection point to represent the maximum of an individual glow peak. This process is iteratively repeated until the remaining area under the original glow curve is reduced below a threshold.

Several additional peak detection methods were investigated during development, including a pure subtraction method which utilized similar concepts as the previously discussed method. The subtraction method however did not utilize a list of local maxima and inflection points; instead the glow curve’s highest point was iteratively determined, calculated an activation energy for the point using a full width at half maximum algorithm, and the peak was individually fit and subtracted from the curve. Also repeated until the resulting curve after subtraction’s area was reduced to a threshold. This method, while partially effective, produced inferior FOM’s when run on the same data as the previously described method on the order of 4-6%. Additionally, the nature of the subtraction method relied on the ascending glow peak maximum intensities, characteristic to the TLD-100. Limiting the software from the more symmetric bell shape the TLD-200.

**2.5. Curve Fitting**

Proceeding glow peak detection, the resulting estimated fitting parameters for the individual peaks are run through a fitting model and undergo a non-linear least square minimization. The software is capable of utilizing the First Order Kinetics model (FOK) described by ([Kitis et al. (1998)](#References)) and the more experimental One Trap Recombination model (OTOR) [(Sadek, 2015)](#References), which will both be discussed in the section to follow. Whichever model is utilized for the fitting is then minimized by an altered iterative Levenberg-Marquardt algorithm (LMA) [(Moré, 1978)](#References), which will additionally be discussed in the sections to follow.

**2.5.1 First Order Kinetics Model**

First described by ([Kitis et al. (1998)](#References)), the fitting equation used to describe a glow curve using first order kinetics was,

*Equation 5 – First Order Kinetics Glow Peak Fitting Equation*

where I(T) is the peak intensity I at temperature T in K, I­­­m is the intensity at the peak maximum, E is the activation energy in eV, k is the Boltzmann constant in eV K-1, Tm is the temperature at the peak maximum in K, is, and is. Each glow peak having its own unique terms E, T, , and for minimization. Each of the individual fitting parameters are represented on an individual glow peak in Figure 5. Out of the fitting models describe by this work the First Order Kinetics model is the model that currently produces the superior results describe in this article. Any others are still subject to research and further development, and more accurately described as experimental.

The equation that the GCA software utilized to calculate the initial guess for the activation energy, E, describe by equation 6 from [(Chen 1969)](#References),

*Equation 6 – Activation Energy*

where is , b is is , where and are the half-width at the low temperature side of the peak and the half-width toward the fall-of the glow peak respectively, is , also depicted in figure 5.

The motivation for exploring alternative fitting models, in particular the OTOR model, is for the improved capability and performance by the software on other TLDs then TLD-100. While the advanced peak detection methods described in section 2.4 are capable of identifying the presence of glow peaks, the First Order Kinetics model runs a lower convergence rate. Results vary in consistency under the model and have thus motivated efforts to explore the OTOR model.

**2.5.2 One Trap One Recombination Model**

The OTOR model was developed to combat a lack for clear physical basis in the general-order kinetic models (GOK) [(Basun et al. 2003),](#References) such as the FOK model described in the previous section. The lack of physical basis inspired the development of more physical “mixed-order” kinetics models (MOK) [(Chen et al. (1981)](#References). The physical basis which describe a MOK model are fundamental basics of the OTOR level model. [(Kitis and Vlachos (2013))](#References) deduced general semi-analytical expressions based on OTOR to describe a single TL glow-peak. The advantages of these expressions that they can accurately describe the TL glow-peaks even in the cases in which the other TL expressions failed [(Sadek et al., 2014a)](#References). Additionally, the OTOR model opens a door to developing non-linear heating rate analytics by the GCA.

The sets of equations which describe the OTOR model require a more rigorous mathematical background, and in turn yielded much more complicated development. Two sets of equations govern the model, determined by the greater of two probability coefficients; the re-trapping probability coefficient, An, and recombination probability coefficients, Am. The two coefficients describe the statistical probability of an electron which has been thermally stimulated from the trap into an excited state either re-trapping or recombining with holes in the centers. The single glow-peak expressions based on the OTOR model are given as [(Kitis and Vlachos, 2013)](#References),

where,

where,

E is the activation energy, k is the Boltzmann constant in eV K-1 ,N is the total concentration of the trapping states in , , , and are the principal and the second branches of the Lambert-W function, respectively [(Olver et al., 2010).](#References)

**2.5.2 Levenberg–Marquardt Algorithm**

The Levenberg–Marquardt algorithm (LM), also known as the damped least-squares (DLS) method, is used to solve non-linear least squares problems. This kind of minimization makes LM a viable choice for solving the GCA software’s curve fitting problem. The software utilizes a modified iterative LM algorithm which minimizes the error between the original emission spectra and the summed glow peak’s curve. Adjusting in parallel parameters, n being the number of glow peaks. The algorithm calculates the error between the two curves, the original and summed, takes the Jacobian, inverts the Jacobian, calculates the Hermitian matrix, multiplies by the identity, and applies a dampening factor to calculate a correction to apply to each individual parameter. This is done iteratively until convergence is met, or failure.

The modified iterative LM algorithm employed by the software on average achieves convergence when run on a TLD-100 within 5-10 iterations, and each iteration in just under 0.3 seconds. The mathematical matrix arithmetic required for the algorithm utilizes the high-speed capabilities of the C language, and utilizes low level optimizations to produce the discussed results at the speeds described. While the complexity of the LM algorithm is difficult to evaluate, [(Bellavia, 2018)](#References) describes the algorithm requiring to obtain an -accurate solution.

**2.6 Output**

Upon completing the deconvolution process the GCA software test the results for convergence, outputs to terminal a list of identified peaks, and their respective curve areas. The deconvoluted peaks are wrote to an output file, placed in an automatically created batch output folder, in CSV format. The output file is easily plotted by using an optional supplemental python script which comes with the GCA package.

Proceeding the final file in the input batch, a batch statistics file is wrote 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. The batch file is intended to allow for analysis of bulk files, and determining trends amongst varying samples.

**3.0 Higher Order TLDs**

The GCA’s ability to branch into higher order TLDs, such as CaF2:Dy (TLD-200) and CaF2:Tm (TLD-300) is still an area of further research and development. Progress with the TLD-200 will most likely rely on the progress of the OTOR model, while less significant development and research may be required to improve TLD-300 deconvolution. Based on the processes described thus far the average FOM for TLD-300 ranges between 5-9%, with convergence rates in the range of 80-90%. Figure 6 depicts a sample deconvolution of a TLD-300 heated at 2.55 degrees per second. The software consistently can identify and estimate peaks, but upon reaching the minimization stage fails to converge to a FOM anywhere near the standard the software sets for the TLD-100. Theoretically the software yields more than capable tools for deconvoluting the TLD-300, however based on the testing data available at the time of testing data noise would appear to play a critical role in GCA’s failure to reproduce the performance of the TLD-100, and further testing may yield positive results.

**4.0 Results**

The majority of the GCA software performance results have been discussed in detail in earlier sections, however as a more detailed overview the results following are based on running large batches of files investigating performance on several varying factors. Varying sample time temperature profiles (TTP), varying irradiation doses , and the peak detection techniques utilized in the deconvolution. The results represented describe the reasons for the choices which were made in the software optimization phase of development.

**4.1 Varying Time Temperature Profiles**

One of the GCA software’s strongest performance results was the testing results across varying TTPs. Testing was run on a batch of roughly 100 properly formatted samples with a varying TTP ranging from 1-10 c/s. The software was able to reach convergence on 99 of the 100 samples, or 99% convergence rate. However, failure to converge was determined to be a result of corrupt data. The test completed in under 4 minutes and achieved an average FOM of 1.6%. What sets the GCA apart from other competitive software is its ability to produce consistent results across all of the varying TTPs, and as figure 7 depicts variation in FOM based on TTP is negligible. A negative slope of 0.08% can be explained by an increase in data noise at the lower TTPs.

An area the project may consider moving forward is the adaptation of non-linear heating rates. Motivation for the ability to deconvolute samples tested under higher order heating rates come from the applications in research that would become available; further research of irradiated materials under varying heating conditions. Further research by the software development team into the mechanics of deconvolution would be necessary, and successful implementation of the OTRO model may be required.

**4.3 Varying Irradiation Dose**

The GCA in addition to superior performance under varying TTPs sets a similar president for varying sample irradiation doses. Prior to heating a TL material is irradiated to a particular dose which is often the subject of the experiment, then during the heating process photons are emitted. The number of photons emitted is proportional to the irradiation dose provided prior to heating. Testing was performed on a batch of 98 samples, as some had to be discarded as blanks and were unable to be replaced prior to testing. The testing similar to earlier tests described was completed in under 4 minutes, yielding a negligibility higher FOM of 1.63%, still far below the expected standard. Irradiation times of the samples varied between 5.5 and 68 minutes, resulting in doses ranging between 2.43-30 mGy. Figure 8 depicts the consistent results produced by the GCA software with a negligible slope of -0.016%.

The software’s ability to produce consistent results across a wide range of sample irradiation was critical to the success of the project. A key concept in thermoluminescent luminescent dosimetry is the vary effects that different doses of irradiation can have on their surroundings, and the software ability to rapidly produce consistent results across large samples of data was a critical feature.

**4.3 Varying Peak Detection Methods**

As described in great depth previously in these findings the experimental implementation of several different automated peak detection methods were explored; most notably two methods, one relying on mathematical derivation and one described as the subtraction method. While both methods yield similar consistency, at least in testing involving the TLD-100, the subtraction method was found to be far more inferior than the method utilizing lists of higher order derivations and has since been discontinued from further development. Figure 9 depicts both the results discussed in section 4.1, testing the TTPs effect on the FOM, and the same test run on a version of the software utilizing the subtraction method. While both methods produce almost identical consistency, FOMs range on average 5% higher when the subtraction method is employed.

A runtime test was preformed, with little to no analysis of the results other than human runtime; just shy of 700 TL spectra were run on the same machine which had had its core resources isolated and configured in an identical way. When both tests were performed the subtraction method concluded in just under 45 minutes, while the method employing mathematical derivations was able to complete the test in almost exactly 38 minutes. This result came as no surprise as the subtraction method required the iterative Levenberg-Marquart algorithm to be initiated n more times than the mathematical method, n being the number of peaks detected. While each of these n instances of the Levenberg-Marquart algorithm were preliminary to the actual minimization stage and required fewer iterations, on the scale of thousands of iterations were able to be avoided without the subtraction method.

In addition to inferior FOM’s and runtimes the decision to move away from the subtraction method is also based on its inability to be developed further into higher TLDs. The nature of the method, and measures which had to be taken to ensure convergence on TLD-100s, disallow for peak detection for glow curves which shape does not share the similar ascending peak shape.

**5.0 Conclusion**

The glow curve analysis software has demonstrated superior performance, runtimes, batch processing, and ease of use than many other popular glow curve software available today. The software eliminates the need for human interaction in the deconvolution process, and in turn produces convergence rates of almost 100%, figure of merits almost half of what the original publication of the figure of merit described as a “good fit” [(Balian and Eddy, 1977)](#References), and the ability to accurately process over 700 thermoluminescent spectra in 38 minutes. The merging of smart computing and the study of thermoluminescent dosimetry have great potential to influence the work being done in the field and help yield more rapidly obtainable and accurate results.

**6.0 Further Work**

Further, and lofty, work that may be considered in the future is employing a machine learning algorithm which automatically identifies the TLD, TTP, a common noise factor for the batch, and irradiated dose, and is able to process samples non-iteratively. Additionally, the adaptation of the OTOR model, while challenging, would open the door to better performance with higher TLDs and non-linear heating rates.

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**Footnotes**

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