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[www.ifj.edu.pl/reports/2005.html](http://www.ifj.edu.pl/reports/2005.html)

Kraków, October 2005

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**Report No. 1968/D**

**The thermoluminescence glow-curve analysis using GlowFit  
- the new powerful tool for deconvolution**

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**Abstract**

A new computer program, GlowFit, for deconvoluting first-order kinetics thermoluminescence (TL) glow-curves has been developed. A non-linear function describing a single glow-peak is fitted to experimental points using the least squares Levenberg-Marquardt method. The main advantage of GlowFit is in its ability to resolve complex TL glow-curves consisting of strongly overlapping peaks, such as those observed in heavily-doped LiF:Mg,Ti (MTT) detectors. This resolution is achieved mainly by setting constraints or by fixing selected parameters. The initial values of the fitted parameters are placed in the so-called *pattern files*. GlowFit is a Microsoft Windows-operated user-friendly program. Its graphic interface enables easy intuitive manipulation of glow-peaks, at the initial stage (parameter initialization) and at the final stage (manual adjustment) of fitting peak parameters to the glow-curves. The program is freely downloadable from the web site [www.ifj.edu.pl/NPP/deconvolution.htm](http://www.ifj.edu.pl/NPP/deconvolution.htm)

## Introduction

Over the last two decades computerized deconvolution of thermoluminescence glow-curves has become the method of choice for TL glow-curve analysis. This method is used to evaluate TL kinetics parameters for a given peak in the glow-curve, but it is also applied nowadays in routine radiation dosimetry. Several models, approximations and minimisation procedures have been investigated and a number of computer programs have been developed for TL glow-curve analysis<sup>(1-5)</sup>. An important step in establishing deconvolution as a reliable research tool was the GLOCANIN project<sup>(6)</sup> – an intercomparison of glow-curve analysis programs. Participants in this project had to analyse TL peak parameters in simulated and experimental glow-curves measured in LiF:Mg,Ti, the most common TL material.

Probably the best deconvolution software currently available is the GCA program developed by CIEMAT<sup>(4)</sup>. This program (version 2.0) has been successfully used in our laboratory over a number of years<sup>(7)</sup>. For well-separated peaks the quality of deconvolution is quite satisfactory. However, the GCA program also has its limitations. A major one is the inability to set constraints on selected parameters. In the case of the newly-developed MTT-type detector featuring enhanced high-LET response<sup>(8)</sup>, constraining some parameters is the only method to obtain meaningful results. Another shortcoming of the GCA program is its manual introduction of initial parameter values, which is time-consuming if large amount of data need to be analysed. Mainly for these reasons, we decided to develop our own deconvolution software, better suited to our particular needs. Within the MS Windows environment, we wished to create a program which could address some of the disadvantages of its predecessors, be more efficient in complex glow-curve analysis, and more user-friendly. We report here a description of the GlowFit program developed by us, and demonstrate results of its performance tests.

## Model and method

The developed glow-curve analysis software is based on the first-order kinetics model of Randall and Wilkins. The TL intensity of a glow-peak in the TL glow-curve is given by the equation<sup>(1)</sup>:

$$I(T) = I_m \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right) \exp\left(-\frac{E}{kT_m^2} \int_{T_m}^T \exp\left(\frac{E}{kT_m} - \frac{E}{kT'}\right) dT'\right) \quad (1)$$

where  $I$  is the glow peak intensity,  $E[eV]$  the activation energy,  $k[eV/K]$  the Boltzmann constant,  $T[K]$  the absolute temperature and  $T_m$  and  $I_m$  are the temperature and the intensity of the maximum, respectively. Because the exponential integral in Equation 1 is not solvable analytically, several different approximations and functions describing a single glow peak, extensively discussed by Bos et al<sup>(2,3)</sup> and Horowitz and Yossian<sup>(1)</sup>, have been proposed. In our case the exponential integral is approximated by the expression<sup>(6,9)</sup>:

$$\int_0^T \exp\left(-\frac{E}{kT'}\right) dT' \approx \frac{E}{k} \int_x^\infty x'^{-2} \exp(-x') dx' = \frac{E}{k} \frac{1}{x} E_2(x) \quad (2)$$

where  $x = \frac{E}{kT} \left(x' = \frac{E}{kT'}\right)$ , and  $E_2(x)$  is the second exponential integral function which can be evaluated by  $E_2(x) = \alpha(x) \exp(-x)$ , where  $\alpha(x)$  is a quotient of 4<sup>th</sup> order polynomials<sup>(9)</sup>:

$$\alpha(x) = 1 - \frac{a_0 + a_1x + a_2x^2 + a_3x^3 + x^4}{b_0 + b_1x + b_2x^2 + b_3x^3 + x^4} \quad (3)$$

Then, finally, the following expression is used to describe a single glow peak<sup>(1)</sup>:

$$I(T) = I_m \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right) \exp\left(\frac{E}{kT_m} \left( \alpha\left(\frac{E}{kT_m}\right) - \frac{T}{T_m} \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right) \alpha\left(\frac{E}{kT}\right) \right)\right) \quad (4)$$

As is shown above, the glow-curve is a non-linear function of  $T_m$ ,  $I_m$  and  $E$  parameters. To find the best-fitted values of peak parameters an iterative procedure must be used. GlowFit is capable of simultaneously deconvoluting as many as ten glow peaks from the TL glow-curve. For ten glow peaks the number of unknown parameters is thirty three - the additional three parameters are used in the background expression, such as  $a + b \cdot \exp\left(\frac{T}{c}\right)$ , where a, b and c are the unknown background parameters. A  $\chi^2$  function must be defined and the best fit parameters are determined by its minimization. The minimization process starts with trial values. In this program the Levenberg-Marquardt method for non-linear function minimization was chosen. In order to enable the user to control the fitting peak parameters, the original algorithm<sup>(10)</sup> was modified to exclude or constrain any given parameter of the peak.

### Program description

GlowFit is a MS Windows-based program and can operate on most PCs. It was written in C++ language. The program is equipped with a user-friendly graphical interface. Typical computer fitting times for an entire TL glow-curve are 1-10 s.

### Starting parameters

Generally, non-linear fitting is strongly dependent on the choice of the initial values of the parameters. The  $\chi^2$  function is much more dependent on the position of the peaks ( $T_m$ ) than on other parameters<sup>(1,11)</sup>, so the first estimation of  $T_m$  is quite critical for reaching convergence. The GCA program uses an automatic algorithm for searching peak positions<sup>(4)</sup>. In principle this may seem to be the best approach, but for some peaks the initially-guessed parameters often require manual adjustment before starting the fitting procedure, which is time-consuming. In the GlowFit another approach was therefore developed. We introduced a so-called *pattern file* which defines the relation between peak parameters and the global maximum of the glow-curve. Some *pattern files* for LiF glow-curves are available together with the program file but if no predefined file is appropriate, the user may create his own *pattern file*. In this case the starting parameters can be easily defined by clicking with a mouse on a plot at positions where peak maxima should be located (energy of 1.7 eV is assumed by default). There is also the option of manual adjustment, which allows the user to easily manipulate each initial value. The typical time necessary for setting up the starting parameters for a new experimental glow-curve consisting of several peaks is usually no more than 1-3 minutes. Once a successful deconvolution is performed a *pattern file* can be automatically generated. Then each next glow-curve of a similar shape can be processed in a few seconds. If the predefined *pattern file* is opened the algorithm starts by finding the global maximum of the glow-curve. Then the temperature distance between position of each peak and the global

maximum are read from the file. The second information read from this file is the ratio between the peak height and the measured TL signal in the temperature channel corresponding to the peak position. Finally, starting values of energy are read from the *pattern file*.

#### Constraints and fixing

The fitting procedure searches for a minimum value of  $\chi^2$  but, unfortunately, this does not imply obtaining ‘true’ peak parameter values. Sometimes the best numerical solution corresponds to unacceptable parameter values. In such cases we should exploit our previously gathered knowledge about peak positions, proportions and energies in order to force the program to maintain the fitted values within preset limits. Such a constraint option was implemented in the GlowFit. The user may set numerical limits or a percentage range around the initial value. Sometimes it is necessary to fix some of the parameters, e.g. in the cases of very complex curves or of very low statistics. The fixed parameters remain unchanged during the fitting procedure. The graphical interface also enables the operator to manipulate the glow peaks after the fitting, as a manual fit adjustment. This may be helpful if the minimization method fails.

#### Results output

Results are presented on the screen and stored in the defined report file. The report contains all the important information: name of the analysed file, a comment (detector type, heating rate etc.), peak height, energy, the temperature position at the peak maximum and the integrals of each fitted peak. In the report a parameter describing the quality of fitting, called Figure Of Merit (FOM) is also stored:

$$FOM[\%] = \frac{\sum_i |y_i - y(x_i)|}{\sum_i y_i} \times 100\%$$

where:  $y_i$  - content of the channel  $i$   
 $y(x_i)$  - value of fitting function in the centre of channel  $i$

Additionally, a plot of residuals, i.e. of the relative differences between measured and fitted values in each channel is also presented on the screen.

The user may also save to the disk a file containing data for plotting all fitted glow-curves and peaks.

#### **Program performance – Reference glow-curves**

As a test, the GlowFit was used to analyze the reference glow-curves from the GLOCANIN intercomparison project<sup>(6)</sup>. In Table 1 the FOM values obtained using GlowFit are compared with the FOM values obtained by a participant of the GLOCANIN, who used the same function to describe a single glow peak and also had one of the best results in the GLOCANIN project (program B in Reference 6). Our results are better in all cases except RefGlow.003 and RefGlow.007.

Table 1. Comparison of FOM [%] values obtained using GlowFit with FOM values obtained within the GLOCANIN intercomparison project [taken from Reference 6].

File name	GlowFit FOM	GLOCANIN Program B <sup>(6)</sup> FOM
RefGlow.001	0.010	0.010
RefGlow.002	0.010	0.010
RefGlow.003	1.180	1.150
RefGlow.004	1.060	1.100
RefGlow.005	1.040	1.070
RefGlow.006	1.230	1.270
RefGlow.007	1.000	0.950
RefGlow.008	0.920	1.130
RefGlow.009	1.350	1.700
RefGlow.010	3.860	4.120

Table 2 shows a comparison between the activation energy obtained by GlowFit analysis with the actual activation energies of the GLOCANIN project for synthetic and average for experimental glow-curves. Agreement for the synthetic glow-curves is within 0.09% or less. These results are rather natural because the data were simulated using the Randall-Wilkins model, the same that was used to develop the GlowFit software. Analysis was performed without any *pattern file* and without setting any constraints. The percentage deviation between the mean activation energies of the GLOCANIN project and activation energies obtained using the GlowFit is below 6%.

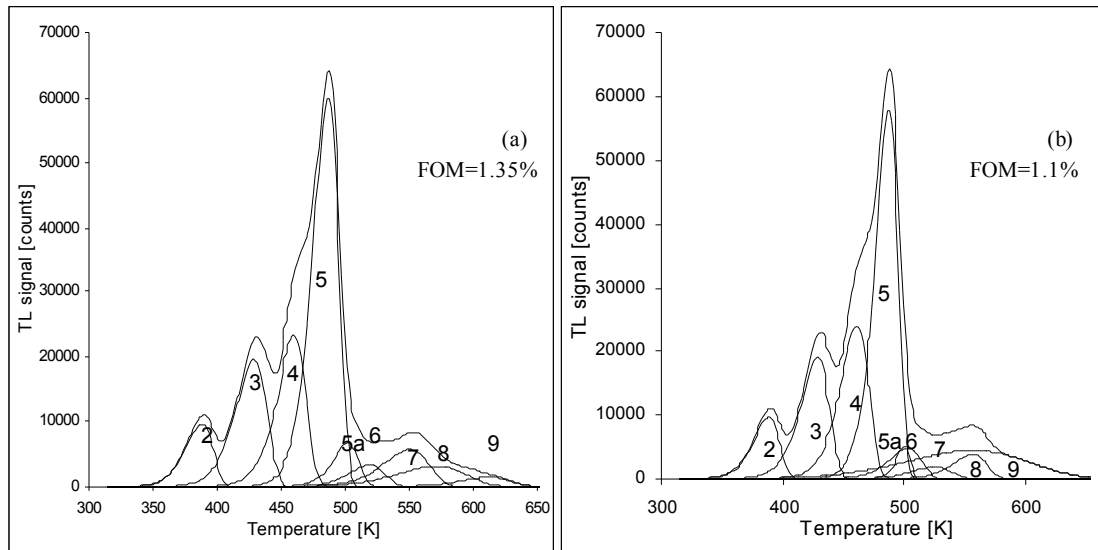
Table 2. Percentage deviation between the activation energies of the GLOCANIN project and activation energies obtained using the GlowFit for synthetic and the experimental reference glow-curves.

File name	Percentage deviation				
	Peak 1	Peak 2	Peak 3	Peak 4	Peak 5
<b>Synthetic reference glow-curves</b>					
RefGlow.001	0.02				
RefGlow.002		0.04	0.05	0.03	0.09
<b>Experimental reference glow-curves</b>					
RefGlow.003		5.6	2.6	1.2	0.8
RefGlow.004		5.7	1.7	2.5	2.4
RefGlow.005		1.2	0.2	3.4	0.6
RefGlow.006		2.4	1.3	2.3	1.3
RefGlow.007			4.2	0.3	1.5
RefGlow.008			1.5	0.6	0.6
RefGlow.009		0.3	0.2	1.3	3.7
RefGlow.010		0.6	1.1	5.9	2.1

The most complicated glow-curve was RefGlow.009, measured for LiF:Mg,Ti irradiated with a dose of 600Gy, which shows a complex structure over high-temperature peak region. These overlapping peaks make the fitting procedure quite difficult. The GLOCANIN participants' results differed not only in the position and shape of resolved peaks but also in their number (9 or 10). Because there are several possibilities of obtaining a good fit, it is not surprising that a great spread, up to 60%, in peak areas and activation energies was found. This complex glow-curve presented in Figure 1 shows how important it is to be able occasionally to introduce parameter constraints to the fit. If peaks strongly overlap, the fitting procedure is evidently not so unequivocal, so information about the peak's position and about other

parameters must be taken from additional experimental sources, e.g. the  $T_m$ - $T_{stop}$  method<sup>(12)</sup>. As shown in Figure 1 (a) and (b), the results are completely different if we analyse this curve with or without constraining the peak parameters. The constrained parameters were  $T_m$  and  $E$  for high-temperature peaks. It should be noted that in both cases the FOM value is very good, and in case of unconstrained fit it is even better, but the physical meaning of some parameters is wrong. Other reference glow-curves were analysed without any parameter constraints.

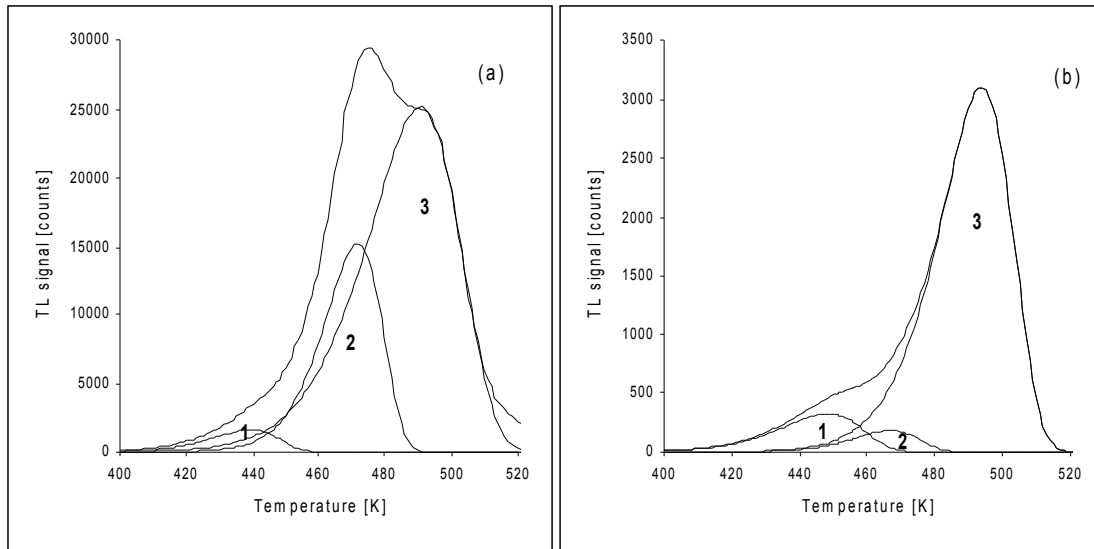
Figure 1. Results of fitting of RefGlow.009 of the GLOCANIN project by GlowFit (a) Fitted with constraining parameters  $T_m$  and  $E$  for high-temperature peaks and (b) fitted without parameter constraints.



### Application of GlowFit for analysis of different LiF glow-curves

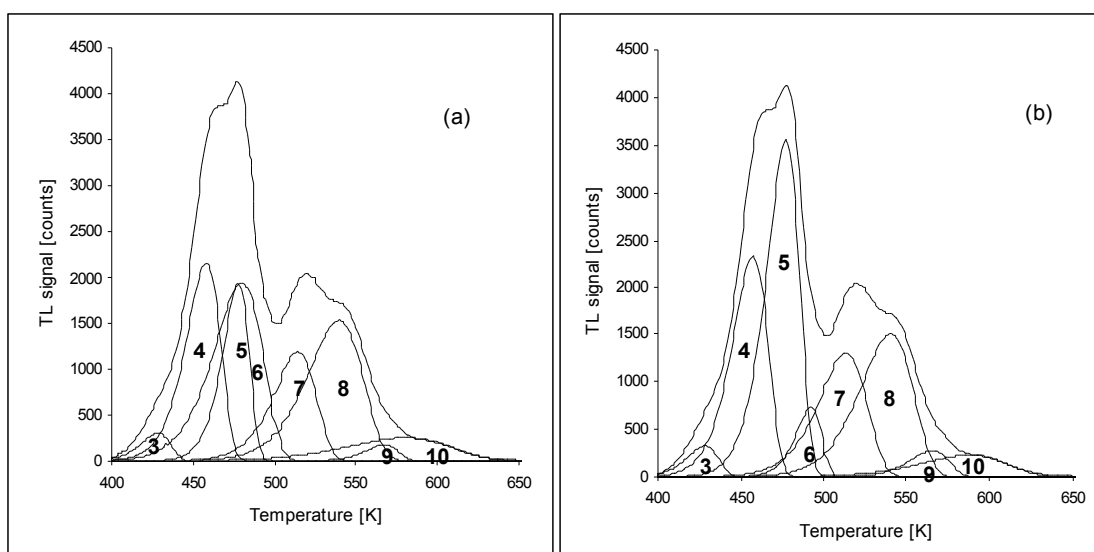
The GlowFit program's main advantage is the ability to analyse complex glow-curves with overlapping peaks. A good example is the analysis of a set of curves measured for an experimental version of LiF:Mg,Cu,P material doped with decreased amount of magnesium (0.05%) exposed to doses up to 1kGy (Fig. 2)<sup>(13,14)</sup>. It was found that at doses higher than 200Gy on the low-temperature side of the main peak a small bulge appears, suggesting the presence of a new peak. At still higher doses this new peak becomes more pronounced. While for the highest doses the deconvolution is quite simple, below 50Gy, when the newly emerging peak is hardly visible or even completely hidden under the main peak, it is impossible to distinguish between the overlapping peaks. From the analysis of curves measured after higher exposures one may determine the position ( $T_m$ ) of this small peak on the low-temperature side of the main peak. According to this knowledge we can limit the peak parameters, especially the temperature of the peak maximum. In Figure 2b we present an example of a deconvoluted glow-curve in this material irradiated by a dose of 49.6Gy, with a small peak appearing at 467K.

Figure 2. Examples of glow-curves in LiF:Mg,Cu,P material doped with decreased amount of Mg (0.05%) irradiated by a dose 467.8Gy (a), 49.6Gy (b), and deconvoluted by GlowFit.



Another good example of the advantages offered by the GlowFit program are MTT-7 detectors irradiated by high doses. MTT-7 is a newly developed version of LiF:Mg,Ti with a changed activator concentration and increased high-LET response<sup>(8)</sup>. Figure 3a presents results of deconvolution using the GCA software (i.e. without constraints), which produced apparently incorrect parameters (peak 6 has moved to the position of peak 5 and an enormously wide peak 10 is observed). When we use GlowFit with constraints set only on the starting parameters of peak 6, which is the most overlapped and difficult to distinguish, the result is such, as shown in Figure 3b.

Figure 3. Example of a glow-curve of MTT-7 material irradiated by a dose of 100Gy; deconvoluted by the GCA program<sup>(4)</sup> (a) and by GlowFit with a constraint regarding peak 6 (b).



## Conclusions

Basing on the successful analysis of GLOCANIN reference glow-curves, we believe that the GlowFit software developed by us is able to accurately and correctly analyse first order TL glow-curves. Unlike similar deconvolution programs, GlowFit is able to resolve complex glow-curves consisting of strongly overlapping peaks, which is achieved mainly through the possibility of setting constraints or fixing any of fitted parameters. Another important feature of the program is the very effective method of setting initial values of the fitted parameters, by using the so-called *pattern files*. GlowFit features a well-developed graphical user interface, which enables easy, intuitive manipulation of glow-peaks in the measured TL glow-curve, prior to the commencement of fitting and after the fit has converged. The GlowFit may be a useful research tool for thermoluminescence studies and for practical dosimetry. It is freely downloadable from the web side [www.ifj.edu.pl/NPP/deconvolution.htm](http://www.ifj.edu.pl/NPP/deconvolution.htm). Further work is in progress to also include the possibility of fitting the second- and general-order kinetics glow-curves.

## Acknowledgments

This work was partly supported by a research project from the Polish State Committee of Scientific Research (KBN) over the years 2003-2005 (No. 4T10C03824).

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# **Appendix A**

## **GlowFit manual**

# 1. Introduction

This software was designed using C++Builder 6 and runs under Windows environment. The best resolution is 1024x768 or higher with small fonts. Distribution disk contains executable file, GlowFit.exe, and two example pattern files.

GlowFit analyses previously recorded measurements of TL glow-curves. Data files should be ASCII files of a maximum of 1000 lines, in the following columns format:

- two columns: temperature (°C) – intensity

The main application window consists of two panels (Figure 1). Operational panel is used to open the data file, set the starting parameters as well as to adjust the parameters after fitting in case the fitting procedure failed. Fitted curve is presented on the screen in the results panel area. The area of each glow peak, FOM value, the residue and number of iterations needed to fit the glow-curve are also shown in this panel.

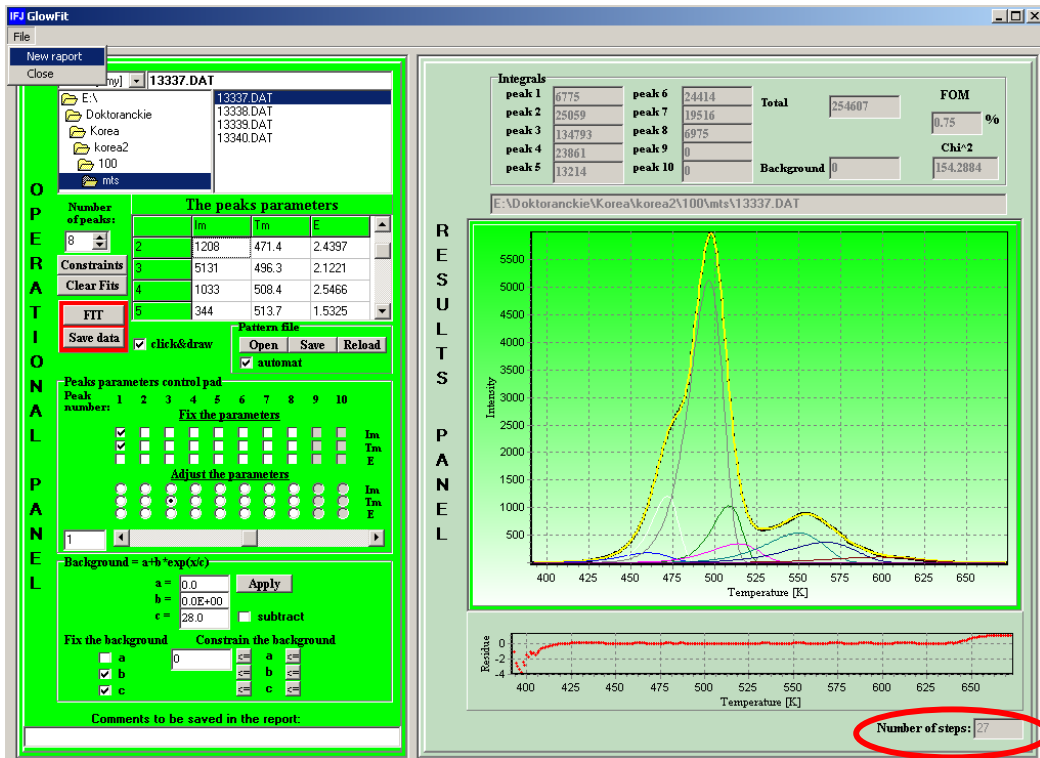


Figure 1. The main application window of the GlowFit software.

## 2. The data file opening

The data file is opened by marking the file name (Figure 2). The plot according to this data appears in the Results panel (Figure 1).

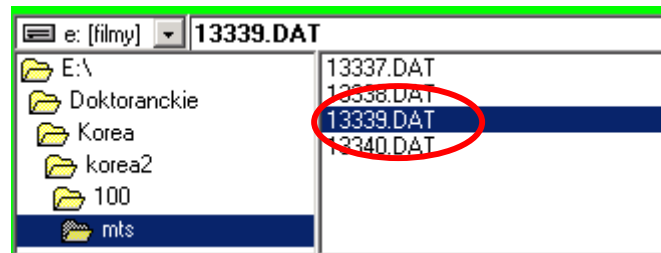


Figure 2. The data file opening.

## 3. The report content

Results are presented on the screen (Figure 1) and also stored in the defined *report* file. At start user should open a new report file (Figure 3) which, after pressing *Save data* button, will contain all important information: name of the analyzed file, any comment (detector type, heating rate etc.), peak height, energy, the temperature position at the peak maximum and the integrals of each fitted peak. In the report a parameter describing the quality of fitting, called Figure Of Merit (FOM) is also stored:

$$FOM[\%] = \frac{\sum_i |y_i - y(x_i)|}{\sum_i y_i} \times 100\%$$

where:  $y_i$  - content of the channel  $i$   
 $y(x_i)$  - value of fitting function in the middle of the channel  $i$

For plotting the fitted glow-curve and particular peaks, the file with .plo extension is saved automatically after using *Save data* button (Figure 3).

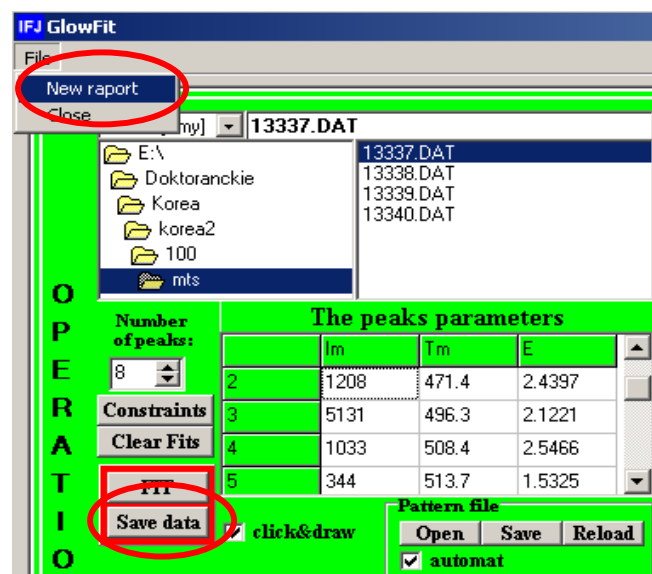


Figure 3. The results storage.

The report and the plot files are the ASCII files, which could be opened by the Notepad or MS-Excel.

## 4. Fitting procedure

### 4.1. The starting peak parameters

There are two possibilities of setting the initial peak parameters.

- **The pattern file**

(*pattern file* is the special file, which contains the coefficients needed to calculate the glow-peak parameters for the typical TL glow-curve. The information read from the pattern file is the temperature distance between position of each peak and the global maximum, ratio between height of a peak and the measured TL signal in the temperature channel corresponding to the peak position. The starting values of energy are also read from the pattern file)

If any pattern file (name.fac) is appropriate to your TL glow-curve:

- ✓ Open it by pressing button *Open* (Figure 4).
- ✓ After opening of a new glow-curve, press the button *Reload* to rescale the peak parameters according to the new global maximum. If checkbox *automat* is marked the reloading is made automatically after opening each new data file.

- **Manual setting**

If none of predefined pattern files is appropriate to your TL glow-curve:

- ✓ Choose the number of peaks (from 1 to 10).
- ✓ Sign the checkbox *click&draw*, which allows you to add the peaks in position within the plot area, where you clicked with the mouse (the default energy value 1.7eV). The initial peak parameters values occur in *The peaks parameters* table.
- ✓ To add the next peak select second row in *The peaks parameters* table and click on the chart again etc.

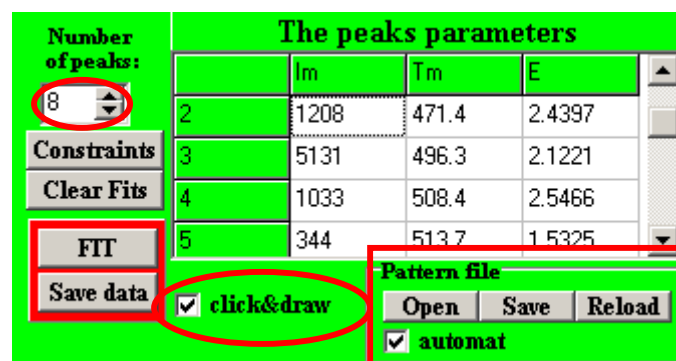


Figure 4. The starting parameters

After setting the initial parameters, button *FIT* must be pressed to start the fitting procedure. Once a successful deconvolution is performed a *pattern file* can be automatically generated by pressing *Save* button. Then each next glow-curve of similar shape can be processed using this pattern file.

## 4.2 Constraints

Sometimes the best numerical solution corresponds to unacceptable parameter values. In such cases user should exploit his previously gathered knowledge about peaks positions, proportions and energies to force the program to keep fitted values within set limits. In GlowFit user may set the numerical limits or percentage range around the initial value. The conditions for the peak parameters may be set by pressing button *Constraints*. To unconstrain the parameter the checkbox of the appropriate peak parameter should be unmarked. By pressing button % user may change the numerical limit into the percentage limit. To back to the numerical limit button <= should be used. The percentage range on all the peaks parameters may be set by pressing button *Set All %*. The once set conditions may be used again, in the next session with GlowFit, after using *Save* and *Open* buttons.



Figure 5. The constraints panel.

### 4.3 The fixing of the peak parameters

Sometimes it is necessary to fix some parameters, e.g. in case of very complex curves or very low statistics. The fixed parameters are not changed during the fitting procedure if user marks the appropriate checkbox as on Figure 6.

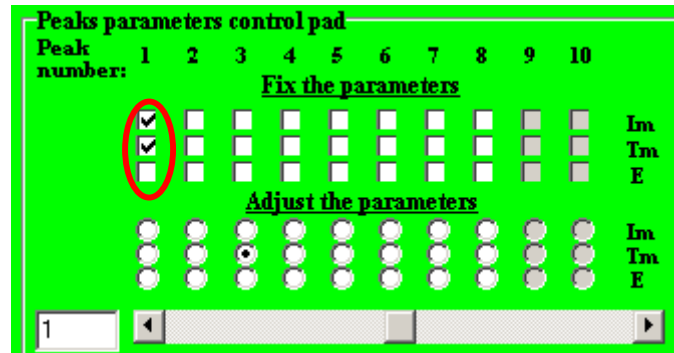


Figure 6. The fixing of the peaks parameters.

### 4.4 The background

The background is set to zero by default. However, user may include the background into the fitting procedure by unmarking fixed background parameters (Figure 7). The background parameters could be constrained by pressing button  $\leq$ . Before starting the fit, the subtraction of background is possible by marking *subtract* checkbox and pressing *Apply* button. This button without *subtract* option plots the background with a, b and c parameters.

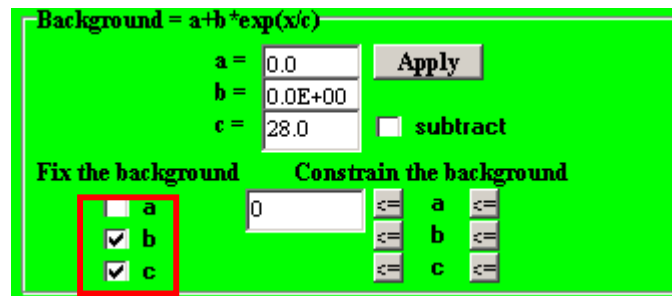


Figure 7. The background settings.

## 5. The fitting quality

A plot of residuals (relative differences between measured and fitted values in each channel) is presented on screen below the fitting curve (Figure 1). Program introduces also the number of iterations needed to obtain the best peak parameters. Number -2 indicates that the maximum number of iterations, 150, wasn't enough to obtain the minimum of  $\chi^2$  function. In this case user should continue the fitting procedure with the new starting parameters, which are the result obtained from the previous fit.

## 5.1 The adjustment

The graphical interface enables also to manipulate the glow peaks before and after the fitting, as a manual fit adjustment. This is very helpful in case the minimization method failed. In order to adjust a peak parameter user should mark the appropriate radiobutton (Figure 8) and move the *slide control* (left(-) or right(+)) to change this parameter value. The step of movement is changeable as shown in Figure 8. Another possibility to adjust the peak parameters is to type the values straightforward into *The peaks parameters* table and confirm by double-pressing **Enter** key (Figure 4).

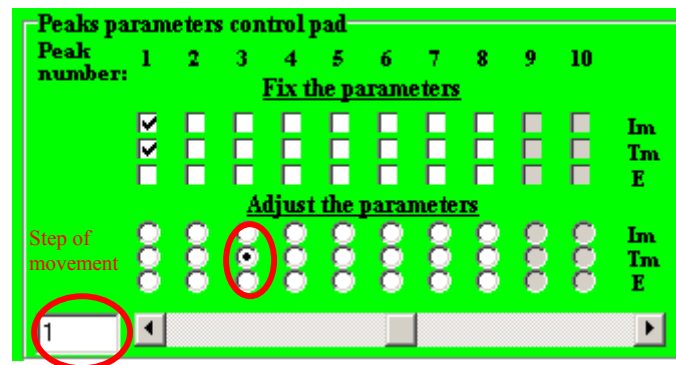


Figure 8. The manual adjustment of the peak parameters.

## 6. Closing

The application can be closed by selecting *Close* from the main menu.

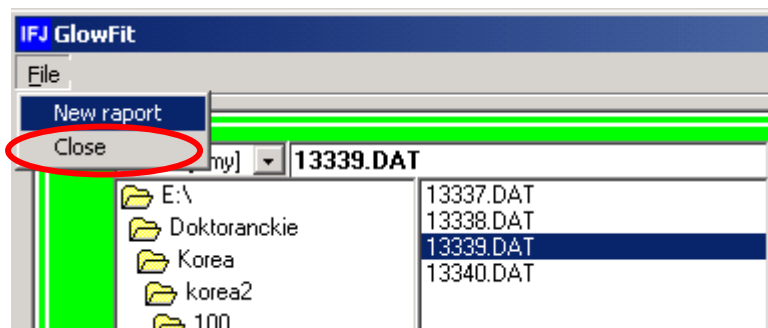


Figure 9. The program closing.



## 7. Overview

<b>Button</b>	<b>Meaning</b>
<b><i>FIT</i></b>	to fit the TL glow-curve (the new window with description DONE indicates the end of the fitting procedure)
<b><i>Constraints</i></b>	to set the constraints on the fitting parameters
<b><i>Clear Fits</i></b>	to clear all set peaks parameters
<b><i>Pattern file</i></b>	<b><i>Open</i></b> to open pattern file – this pattern will be valid till opening the new one
	<b><i>Reload</i></b> to rescale the peak parameters according to the curve global maximum (do it after opening each new curve or mark <i>automat</i> to do it automatically)
	<b><i>Save</i></b> to save the pattern file
<b><i>Save data</i></b>	to save the important data to the report file and the plot coordinates to filename.plo file
<b><i>Apply</i></b>	to show the background with a, b, c parameters (when <i>subtract</i> is checked the background will be taken away from the data source)
<b><i>double-click on the chart</i></b>	to change the linear scale into the logarithmic (to back into linear scale double-click again)

## 8. Errors

In case of any error, please restart the program and be so kind to send e-mail to author describing the problem.

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