Journal of the Korean Nuclear Society
Volume 29, Number 5, pp. 385~392, October 1997

The Retrieval of Abnormal TL Glow Curves Using Modified Glow Curve Analysis Method

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(Received February 4, 1997)

Abstract

The shape of TL glow curve is a useful indicator for assurance of correct reading of the personal dosimeter. Since the reading procedure of TLD is irreversible, however, an analytic remedy should be considered to procure reliable dosimetric information for the readings with irregular glow curve shape. In this study, kinetic trapping parameters of $CaSO_4$: Dy Teflon personal dosimeter(Teledyne PB-6A) were analyzed by Halperin and Braner's model for general-order kinetics. From these kinetic trapping parameters, we also developed a simple procedure to retrieve the dosimetric information from abnormally distorted glow curves. The computerized glow curve deconvolution(CGCD) fitting of the reference glow curve with kinetic parameters from this study yields relative errors of about 5% from the expected integral. It was also found that the glow curve remedial procedure developed could retrieve the distorted TL glow curves within error ranges of 15%. With the glow curve retrieval techniques, doses incurred by gamma radiation can now be succefully re-constructed for the $CaSO_4$: Dy Teflon dosimeter resulting abnormal glow curves.

1. Introduction

Applications of computerized glow curve deconvolution(CGCD) to thermoluminescence dosimetry (TLD) began to appear in the scientific literature in the early 1980s, not far behind the introduction of personal computers into academia and industry. It was fairly obvious that the application of well-known mathematical fitting routines to the glow curve of most common TLD materials might lead to important advances in TLD. Earlier disadvantages due to computer speed limitations have largely been resolved. CGCD can be carried out in real time as the individual dosimeters are read out, and iteration times on the modern generation of personal computers with mathematical coprocessors(up to speeds of 200 MHz) are measured from milliseconds to fractions of a second.

The quality control of a TL dosimetry system is potentially one of the most important applications of CGCD. One can envisage important material selection improvements in the manufacturing stage of the

material, in the manufacturer's quality control of the final product, and in the final stage of glow curve readout involving verification of dose data as well as diagnostics of reader malfunction[1]. The final stage is especially important since glow curve analysis automatically involves a form of permanent storage of the dose information and completely answers one of the complaints in the use of TLD regarding a permanent record of the dose information. Lucas[2] may have been the first to suggest the great potential of glow curve analysis in the verification of data. He listed a number of faults that could be recognized automatically through computer. These include : (i) absence of glow peaks, (ii) failure to heat the chip, (iii) electrical interference, (iv) a glow peak at the wrong temperature, (v) saturation of the readout, and (vi) a high dark current (due to a light leak or other electric malfunction). Hoot and Landrum[3] in 1982 were the first to compile and publish a library of various error conditions and to suggest that the dose reading could be verified by glow curve analysis. These abnormal dose readings could be very significant as shown in Table 1.

The purpose of this study is to develop a computational method for the retrieval of abnormally distorted glow curves. A computerized analysis was performed on a measured glow curve originating from the well-developed TL material $CaSO_4$: Dy (Teledyne PB-6A). The analysis method chosen was based on unfolding the TL glow curve into sub-peaks using Halperin and Braner's equation for general-order kinetics[4]. The Levenberg-Marquardt fitting method and striping procedure were also applied for non-linear curve fitting of pre-determined parameters to experimental data[5]. From the results of trapping parameter analysis, a simple mathmatical procedure was drived to approximate the initial charged particle concentration of distorted sub-peak. In the next, the applicability of the retrival method to a distorted glow curve was tested with arbitrary distorted curves and the analysis of retrieved results were given by the integration of interesting region.

Table 1. Evaluated Dose Readings for TL from Abnormal Situation

Contaminant/external agent	mrem equivalent
2min to a TLD exposed to 1R ^{137}Cs	919
Anti-static spray	17
WD-40	13
Chalk dust	36
Soaps	35
Dirt from floor	92
Plastic slivers	27
Oil and hand lotion	39
Electric sparks(>30kV for 10sec)	2995

2. Theory

Although different equations are in use to describe a thermally activated process, a common description is given by Eq.(1) which describes the rate of release $I(t)$ of some kind of particles initially trapped by a number $n(t)$ of traps and released by thermal activation as a function of time t with initial condition $n(t=0)=n_0$[6].

$$I(t) = -\frac{dn}{dt} = s' n^p(t) e^{-E/kT} \tag{1}$$

Here E is the energy needed to release a particle from a trap, s' the attempt-to-escape frequency, k the Boltzmann constant, and p the kinetic order. The temperature T is varied in a controlled way as a function of time t.

A solution of Eq. (1) can be obtained by dividing by n^p and direct integration with $s = s' n_0^{p-1}$.

$$n(t) = n_0 [1 + (p-1) s \int_0 e^{-E/kT(t')} dt']^{-1/(p-1)} \tag{2}$$

Generally, the rate of release of particles is measured with some efficiency ε with $m_0 = \varepsilon n_0$.

$$J(t) = \varepsilon I(t) = -\varepsilon \frac{dn}{dt}$$

$$= m_0 s e^{-E/kT} \left(1 + (p-1) s \int_0 e^{-E/kT'} dt'\right)^{-p/(p-1)} \tag{3}$$

For the hyperbolic heating temperature profile, Eq. (3) can be accurately approximated as follows[7] ;

$$J(t) = m_0 \frac{bE}{k} e^{(bE/k)(t-t_m)} \left[1+(p-1)e^{(bE/k)(t-t_m)}\right]^{-p/(p-1)}$$
$$= J_m e^{(bE/k)(t-t_m)} \left(\frac{\left[1+(p-1)e^{(bE/k)(t-t_m)}\right]}{p} \right)^{-p/(p-1)} \quad (4)$$

where, index m referring to the quantity at peak maximum and $1/T(t) = 1/T_0 - bt$ with T_0 the starting temperature and b a coefficient determinig the heating rate.

In order to be able to fit parameters m_0, s, and E from one or more theoretical glow peaks to a measured composite glow curve, the number of peaks must be determined. Some smoothing procedure was applied before determining peak position. At this stage, peaks were only taken into account if the intensity on either side of the peak drops to less then 90% of the peak's maximum intensity. Next, the kinetic order of each peak was determined. As the asymmetry of the peak is closely related to the kinetic order, the asymmetry parameter α was calculated at various fractions γ of the maximum intensity and the quantity $Q(\gamma)$ defined by Eq.(5) was considered [7].

$$Q(\gamma) = \frac{(1-\alpha)^2}{(1-\gamma)} \quad (5)$$

$$\alpha(\gamma) = \frac{t_R - t_m}{t_m - t_L} = -\frac{ln\, z_R}{ln\, z_L} \quad (6)$$

$$z_{L,R} = z(t_{L,R}) = e^{(bE/k)(t_{L,R}-t_m)} \quad (7)$$

Once the kinetic order is determined, the energy parameter E can be estimated as follows :

$$E = \frac{k}{b} \left| \frac{ln\, z_{R,L}(\gamma)}{t_m - t_{R,L}(\gamma)} \right| \quad (8)$$

Here, either Z_R or Z_L can be used when the corresponding value is taken. Having determined E, the parameter s can be obtained from Eq.(9) which holds accurately for any kinetic order and temperature profile.

$$s \approx (bE)e^{E/kT_m} \quad (9)$$

This approximation is very accurate if $exp(-E/kT_0) \ll exp(-E/kT)$. This condition will always be fulfilled in practical situations, where $T_0 \ll T_m$ and J significantly differs from zero only if T is close to T_m. We shall therefore no longer denotes this as an approximation.

Finally, the parameter m_0 can be estimated from Eq. (10) for hyperbolic temperature profiles.

$$m_0 = (k/bE)J_m\left[p-(p-1)\right] \times \left(1-\left[(p-1)/p\right]\right)^{-p/(p-1)} \quad (10)$$

After initial estimates of all the parameters of a peak conducted, the contribution of these peaks to the total glow curve was calculated. When all peaks are finally identified and their parameters estimated, we have applied the non-linear least-square method to search the minimum of a multi-dimensional function ;

$$\chi^2 = \sum_T \frac{\left[N^{exp}(t) - G(t)\right]^2}{\sigma^2(t)} \quad (11)$$

with $N^{exp}(t)$ the experimental value of TL light at time t, $G(t)$ the expected value of $N^{exp}(t)$ with predetermined kinetic parameters, σ^2 the variance of expected value. The experimental data counts $N^{exp}(t)$ are distributed as Poissonian variables with a mean value $\mu \geq 10$. In this case the Poissonian variable could be approximated by a Gaussian variable and the least square method is applicable.

3. Experimental

In order to determine the trap parameters of a reference TL glow curve, a computerized analysis was performed on a measured glow curve originating from the well-developed TL material $CaSO_4$: Dy (Teledyne PB-6* , Teflon Disc-type). The TL material used has an active diameter of 7.9mm and a thick-

* Teledyne-Brown Engineering Co.

ness of 0.4mm. Prior to the irradiation, the sample dosimeter was annealed at 260℃ for 2 hours and then cooled down naturally to room temperature. Irradiation was performed at the national secondary dosimetry laboratory in Korea Atomic Energy Research Institute with a standard ^{137}Cs source to the dose levels of 1.5 Gy. The sample was read out about 24 hours after the irradiation for considering dosimeter fading effect. To suppress tribo-thermoluminescence and chemi-thermoluminescence, dry (<2 PPM H_2O) ultra-pure N_2 was used (transported to the reader via PTFE tubing) at a flow rate of approximately 50 cm^3/sec. Glow curves were obtained using a Teledyne A300 that was interfaced via an GPIB port to an 486DX2-50 personal computer used to perform the in-situ glow data acquisition. A typical glow curve of ^{137}Cs gamma ray irradiated $CaSO_4$: Dy chip is shown in Figure 1. At the heating rate employed, the maximum intensity of peak occurs at about channel 80 (a temperature of approximately 220℃) and it shows a considerable agreement with previous works[1, 8]. The initial peak range (~65 channel) of glow peaks in Figure 1 also contains the background signal attributable to electronic noise and infrared emission from the heating block and chip itself.

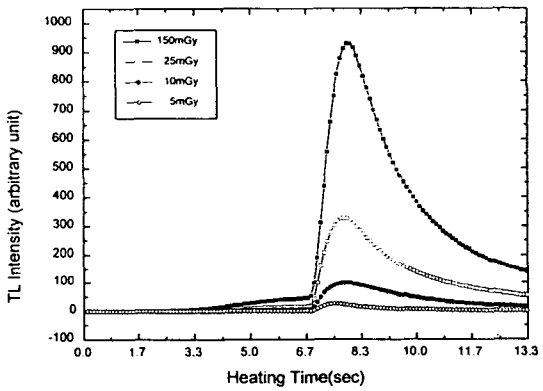

Fig. 1. Glow Curves from a Teledyne PB-6A $CaSO_4$: Dy Teflon for Various Dose Levels

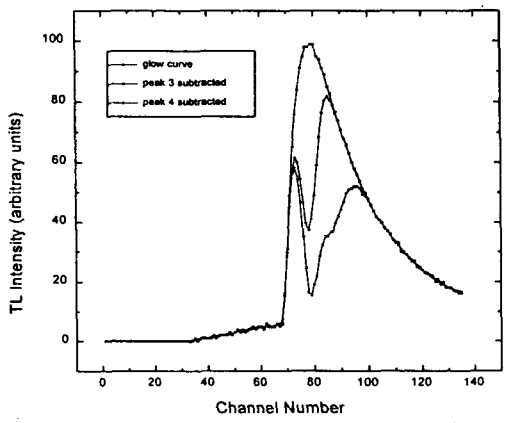

Fig. 2. Glow Curve Obtained from a Dosemeter Irradiated with 20mGy Gamma and the Same Glow Curve with Sub-Peaks Subtraction.

4. Results and Discussion

4.1. Kinetic Parameter Analysis

To obtain good initial estimates in the first search for peaks, first dominent peak(number 3) was taken into account and its parameters estimated. From the residual glow curve after subtraction of the calculated contribution of peak 3, peak 4 was identified and its parameters estimated, and so on. In Figure 2, it can be seen that the result of primary peak substraction from glow curve gives an identification of residual peaks. The Levenberg-Marquardt iteration method and striping procedure was applied for non-linear curve fitting of pre-determined parameters to experimental data[5]. With the peak symmetry analysis with Eq. (5), Q values as a function of γ indicated beyond second-order kinetics for all six peaks. It is usually assumed that $1 \leq p \leq 2$, but TL peaks with values of p beyond this range have also been found. It is difficult, however, to envisage a physical model for which $p > 2$[9]. We therefore treated the kinetic order in this study as a non-free parameter. The resulting initial estimates and final estimates of the peak parameters are shown in Table 2. A fit with the six identified peaks resulted from the final estimates of the peak parameters is also shown in Figure 3 which are presented in 135 equal-width channels. The sum of

Table 2. Parameters of the TL Glow Peaks in $CaSO_4$: Dy(PB-6A) Teflon Obtained by Computerized Glow Curve Deconvolution.

		Peak Number					
	Parameter	1	2	3	4	5	6
Initial Estimates (χ^2 =0.807)	$t^m(sec)$	5.857	7.275	7.802	8.460	9.367	11.493
	J_m	3.774	52.728	61.428	46.236	35.692	23.518
	order	2	2	2	2	2	2
	E(eV)	1.067	1.160	1.713	1.756	1.122	1.226
	$s(sec^{-1})$	7.29E14	1.11E14	9.86E18	2.01E18	9.97E10	1.28E11
	m_0	11.77	28.91	50.51	65.15	100.06	138.88
Final Estimates (χ^2 =0.059)	$t^m(sec)$	5.910	7.252	7.728	8.441	9.579	11.877
	J_m	3.916	41.250	63.855	56.981	37.953	22.604
	order	2	2	2	2	2	2
	E(eV)	1.053	1.114	1.343	1.573	1.478	1.817
	$s(sec^{-1})$	4.27E14	3.31E13	9.76E14	2.55E16	2.85E14	4.30E16
	m_0	12.37	22.56	58.84	86.74	99.59	108.50

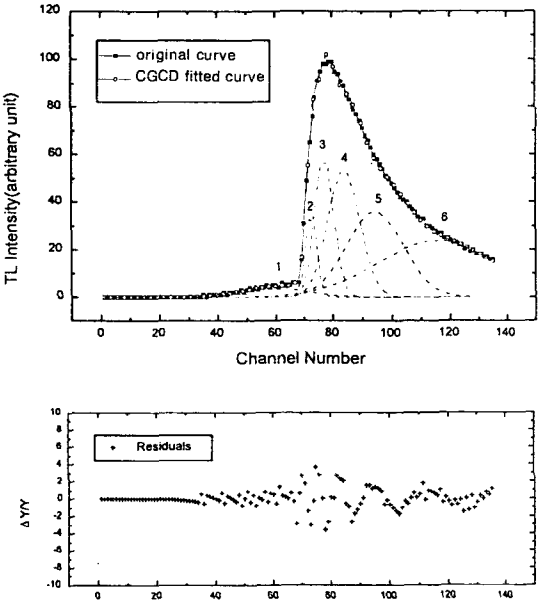

Fig. 3. Deconvoluted Glow Curves for a Teledyne PB-6A $CaSO_4$: Dy Teflon by CGCD.

the deconvoluted glow peaks is also compared to the experimental glow curves in Figure 3 which shows the residuals ΔY/Y, plotted versus chennel, where

$$\frac{\Delta Y}{Y} = \frac{Y_{experimental} - Y_{fitted}}{Y_{experimental}}.$$

4.2. Retrieval of Distorted Glow Curve

From the deconvolution results of Table 2, it can be noted that the concentration of initially trapped electrons(m_0) shows monotonous increasing trend with the peak center time t_m. In general, it should be noted that the shape and peak temperature of the fir st-order glow curves are independent of the initial concentration of the trapped charge while, in contrast, both the shape and peak temperature of the second-order glow peaks may(or even usually) vary appreciably with initial trapped charge, i.e. with dose [10]. The initial concentration of charge m_0, therefore, is a essential parameter for characterizing the property of glow curve. As illustrated in Figure 4, this relationship between the initial charge concentration and peak center time was also found for various dose ranges. As mentioned above section, kinetic trapping parameters E and s of sub-peaks are the unique value for the same TL signal acquisition environment(i.e. materia and heating profile) and can be considered as independent parameter. We can, therefore, develope the simple procedure to approximate the parameter m_0 of distorted portion of glow curve from that of undistorted portion.

From Eq.(4), the maximum intensity of peak J_m for

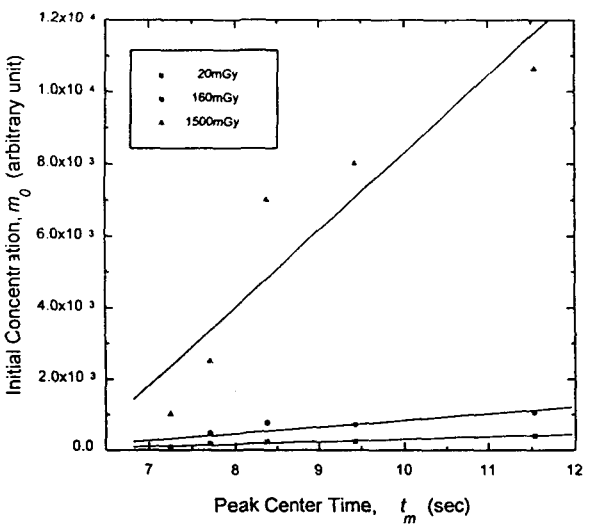

Fig. 4. The Response of Initially Trapped Charge Concentration as a Function of Peak's Center Time for Various Dose Levels.

the hyperbolic temperature profile can be rewritten as follows.

$$J_m = m_0 s e^{-E/kT_m} p^{-p/(p-1)} = (m_0 bE/k) p^{-p/(p-1)} \quad (13)$$

From Eq.(13) , it can be known that if we can get the maximum intensity J_m of some glow peak, then we can guess the initial concentration m_0 of that peak since the other parameters are constant and already known. As mentioned above, the concentration of initially trapped electrons shows somewhat linear response to the peak center time m_0 for various dose levels. After the initial guess of concentrations of undistorted peaks conducted, therefore, we can obtain the other peak's parameter m_0 of distorted portion using a general linear regression method.

In order to test the applicability of this glow curve retrieval procedure, application to two kinds of synthetically distorted glow curves of PB-6A TL material exposed to different doses of gamma radiation was investigated. The analysis of the glow curve was carried out by predicting the initial parameters from undistorted peaks(peak1 and peak6). In this particular exercise, and for demonstration purposes, identification of undistorted sub-peaks was carried out by visual inspection and mathematics package software [11]. It is, however, no problem to configure the fitting procedure automatically to identify and analysis the distorted region. A typical result of the computerized analysis and comparison with the undistorted

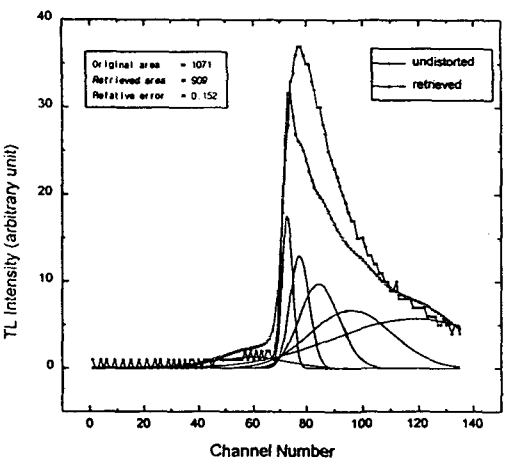

Fig. 5. Retrieval of Distorted Glow Curve Obtained from a Dosemeter Irradiated with 3mGy Gamma by Initially Trapped Charge Concentraion Approximation.

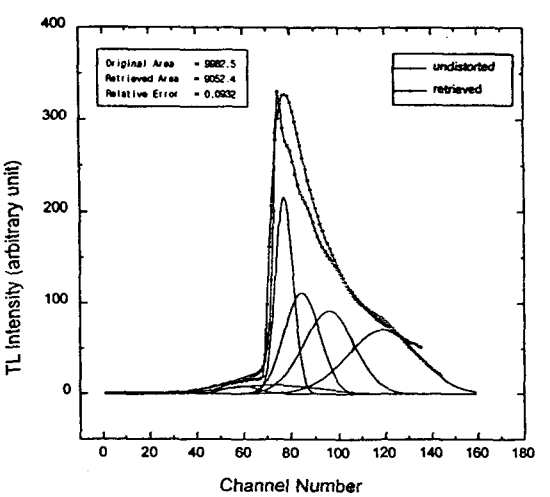

Fig. 6. Retrieval of Distorted Glow Curve Obtained from a Dosemeter Irradiated with 22mGy Gamma by Initially Trapped Charge Concentration Approximation.

Table 3. Retrieved Kinetic Trapping Parameters of $CaSO_4$: Dy Glow Curve Irradiated with 3mGy Gamma Rays.

Peak	Trapping Parameter			Retrieved Parameter		Relative
Number	t^m(sec)	E(eV)	s(sec^{-1})	J_m	m_0	Error
1	5.910	1.053	4.27E14	2.0	6.32	
2	7.252	1.114	3.31E13		11.63*	0.152
3	7.728	1.343	9.76E14		13,85*	
4	8.441	1.573	2.55E16		17.15*	
5	9.579	1.478	2.85E14		22.38*	
6	11.878	1.817	4.30E16	7.0	33.60	

* Values are approximated by linear regression.

Table 4. Retrieved Kinetic Trapping Parameters of $CaSO_4$: Dy Glow Curve Irradiated with 22mGy Gamma Rays.

Peak	Trapping Parameter			Retrieved Parameter		Relative
Number	t^m(sec)	E(eV)	s(sec^{-1})	J_m	m_0	Error
1	5.910	1.053	4.27E14	15.0	47.40	
2	7.252	1.114	3.31E13		119.75*	0.0932
3	7.728	1.343	9.76E14		145.58*	
4	8.441	1.573	2.55E16		183.91*	
5	9.579	1.478	2.8E14		244.69*	
6	11.878	1.817	4.30E16	77.0	367.98	

*Values are approximated by linear regression.

glow curve are shown in Figure 5, 6 and related parameters are summarized in Table 3.

As seen from Table 3 and 4, glow curves retrieved from the finally estimated parameters follow closely the measured glow curves with the error ranges of 0.09~0.15. When this procedure was expanded to the experimental glow curves from various dose levels, it was seen that computerized retrieval fitting of the glow curve yields a difference of the area integral within about 20% from the expected integral. The accuracy of the retrieved information depends on the extent of the damage to the glow curve.

5. Conclusions

A new procedure to identify and retrieve the distorted components in $CaSO_4$: Dy (PB-6A) glow curves have been presented. This procedure has been implemented in a rather simple computer program to analyze these curves and retrieve the damaged components necessary for dose reassessment. It would seem that if curves are captured during the processing steps, some quantitative analysis, no matter how slight, is important to give additional assurance that the processing of an individual in-field dosimeter resulted in the correct interpretation of the data. The use of this procedure has substantially improved Teledyne TLD system A-300 dose re-estimation capability by reliably extending the workable situation. Future efforts will be directed at a study of the accuracy of the retrieved information as a function of the temperature range over which the distortion occurs.

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