

Preface

One may perhaps question the need for another book on this subject when so many of them have been written in the past.

The title of the book—"Unravelling Thermoluminescence" in some way provides an answer to this query. The dictionary defines "unravel" as an attempt to "make plain or clear; solve or explain something complicated or puzzling." That is what this book seeks to do.

The first two chapters are written assuming that the reader is a fresh undergraduate who is beginning to take up work in the field of thermoluminescence (TL). The subject of TL, being one among the variety of luminescence phenomena in nature, it is thought fit to acquaint the student with these various phenomena.

Chapter 1 includes the historical development of the subject of TL and also gives a brief overview of the luminescence phenomena.

Chapter 2 summarizes well-known types of point defects in the inorganic solids. These serve as simple examples of defect centres, the types of which may act as traps and recombination centers. However, the actual structure of the defect centers in different TL phosphors is not simple to find out, neither is it necessary for the application of TL in areas like dosimetry and archeological and geological dating. X-rays and nuclear radiations being the principal sources which induce the TL, their interaction with matter is summarized in this chapter. The interactions are discussed in the perspective of the TL induction by different types of radiations.

The subsequent chapters go on to address those points that are new or those that have not been included in the books published earlier. Such parts have been highlighted by showing them in italics. The stress has been more on explaining the concepts than reviewing the earlier works. In the process reference to earlier literature is not comprehensive.

Chapter 3 begins with the explanation of the thermal stimulation process and goes on to describe conventional models of TL. The characteristic properties of the glow curves of the Randall–Wilkins, the Garlick–Gibson, the general order (GO), and the mixed order (MO) kinetics models are summarized. *While dealing with the GO kinetics model it is shown that the pre-exponential factor s' and the kinetic order (KO) parameter b are not independent constants as assumed by the*

proponents of the model, but that the s' value is dependent on the value of b and the total concentration N of the traps. When GO kinetics model is applied to the simple one trap one recombination center (OTOR) physical model, it is seen that the b value itself varies with n_0/N , which means it varies with dose given to the sample. The lacunae in GO kinetics model are discussed in Sects. 3.3.4 and 3.3.4.1. The MO kinetics model too has drawbacks. A major drawback of MO model is that its KO parameter α is dependent on n_0 , which means that in real samples it would depend on the dose given to the sample. Thus no unique value of α may be assigned to a given glow peak of a sample. The chapter then takes up the physical models. These include the multi-trap systems which are the simplified version of the generalized scheme consisting of a host of traps and an equally large number of recombination centers. These may plausibly be applicable to the real materials. It is shown by simulations how first order (FO) kinetics glow peaks are produced under a variety of parametric conditions. The results of these simulations are used to answer the question why the KO of the TL glow peaks of real materials is invariably seen to be of FO. The conclusion drawn is that there is an abundance of thermally disconnected deep traps which provide for the recombination rate to exceed the retrapping rate and thus produce FO kinetics. Further, at low trap occupancies, FO is produced even in retrapping dominant conditions. It is logical that defects would exist in a crystalline material until it approaches its melting point. Examples are given of some materials in which deep traps have been detected. Another way by which FO kinetics is produced is the local recombination of the thermally excited charge carriers. Examples are given from the published papers wherein local recombination is proved.

Chapter 4 deals with the kinetics analysis of the TL glow peaks. Five methods of glow curve analysis are summarized including the merits and the demerits of each method and the precaution in their application. The parts which I may call worth mentioning as new in this chapter are as follows:

- (1) A stable peak temperature T_m irrespective of trap occupancy n_0/N (dose) is the property exclusive to the FO kinetics. In all non-FO cases the glow peak temperature changes with trap occupancy. This property may be used to differentiate the non-FO cases if any, from the FO ones before attempting the kinetics analysis of any experimental glow curve which requires kinetic order. Application of peak shape method in analyzing the experimental glow curves may face uncertainty in the shape due to the presence of weak satellites close to the peak under study.
- (2) The peak temperature of the simulated TL glow peaks is found to vary linearly with the activation energy E for a given s/β . Examples of real samples are given which also show that activation energy E of different glow peaks of the given sample are almost linearly related to the glow peak temperature. This implies that the value of the frequency factor s for the different glow peaks of a given sample may be same.

- (3) *A method has been suggested to find the value of KO using the initial rise profiles. It is a new method which has been proposed for the first time. The method is general in the sense that it is applicable to all values of KO.*
- (4) *If a GO kinetics glow curve is analyzed by using the isothermal decay method, it turns out that the resulting E and the KO values are dependent on the initial concentration n_0 of the active traps (dose). This result once again brings out the lacunae in the GO kinetics expression.*
- (5) *Application of the peak fitting method to the simulated glow peaks of different physical models shows that the error in the found value of E and the minimum error in fitting vary with the best fitted value of b. The found value of E turns out to be error free only when the corresponding best fitted value of b is either 1 or 2. For cases in which the best fit value of b lies between 1 and 2, error in found E value is seen to be highest when the value of b is 1.5. At this point the error value also is seen to be highest. The highest error in found value of E at this point is about $\pm 8\%$. For best fit b values less than 1 or more than 2, error in found value of E are much higher and so are the minimized fitting error values.*
- (6) *An example taken from a published paper on post-irradiation annealed LiF: Mg, Ti sample shows that the obtained values of E as well as KO using the curve fitting method increase with the annealing time. When we examine the analysis procedure used by the authors of the paper we see that the minimum value of the error function (FOM) also has increased with the annealing time. This implies that the minimization of the error function in curve fitting method does not ensure the accuracy in the obtained value of E if the best fit value of KO is found to be different from 1 or 2.*

Chapter 5 deals with the quasi-equilibrium (QE) problem in TL. To the knowledge of this author a large part of the material of this chapter has not appeared in earlier books in this field. *It is concluded in certain earlier publications that QE cannot be achieved under the condition in which retrapping rate is much faster than the recombination rate. In this chapter this conclusion is disproved and it is shown that it is the values of the system parameters, namely the concentration of the traps and the recombination centers and their cross sections, which determine whether or not the QE condition would be satisfied. In this connection the importance of the recombination lifetime is highlighted. Smaller the value of the recombination lifetime, better it is for the QE to be achieved. Using the premise that under the QE condition the relaxation rate of the thermally excited carriers should be equal to the excitation rate, the minimum value of the function $\sum_i N_i \sigma_i$ for the validity of the QE approximation is found, where N_i stands for the concentrations of the active traps and the deeper traps and σ_i stands for their respective cross sections. This is a new approach to find the range of parameter values to satisfy the QE condition.*

It is shown for the first time that the QE depends not only on the parameter values of the system but also on the heating rate. At a certain high heating rate, depending on the parameter values of the system, the QE begins to deteriorate, which reflects as change in the peak shape. It is suggested that the heating rate dependence of QE may be used to test whether or not QE approximation holds for an experimental glow curve. It is shown that the heating rate dependence of QE may be used also to find the range of the values of parameters (trap populations and the cross sections) at least approximately, for an experimental glow peak. As an illustration this test is applied to the glow peak No. 5 of LiF TLD phosphor.

Chapter 6 deals with the growth of TL intensity with radiation dose. After a brief review of earlier theories and earlier works on this subject, the discussion takes up a model which gives physical meaning to the so-called competitors during the TL read out heating. *As against the arbitrarily assumed competitors by the earlier workers, in the proposed model the competitors are the thermally disconnected deep traps (TDDT) which when empty act as competitors and when filled cause increase in the number of recombination centres.* When the active traps and the TDDTs get filled up during irradiation the reduction in competition and the increase in filled active trap population take place simultaneously. As a result the TL intensity growth becomes superlinear. The treatment of the model takes into count the irradiation and the heating stages together. *Using this model expressions are derived for the TL intensity growth curve, the supralinearity factor (SF), and the pre-dose sensitization factor (PDSF). Also derived is a new expression called the sensitization factor (S_nF). The factor S_nF which has been introduced by this author is used to give a unified explanation for the mechanisms which give rise to the SF and the PDSF. The computed profiles of SF, PDSF, and S_nF are compared with the experimentally obtained profiles of these factors for the case of the LiF:Mg, Ti TLD phosphor. These are found to match well with each other supporting the soundness of the model used.* Apart from providing a theoretical basis underlying the phenomena of supralinearity and pre-dose sensitization, the model dispels the doubt of some workers that the mechanism involved in these two phenomena may be altogether different from each other.

Chapter 7 deals with photo-transfer of TL (PTTL) intensity from one glow peak to another. This phenomenon is observed almost in all thermoluminescent materials, usually showing a transfer from higher temperature glow peak to those of lower temperatures. *In this chapter transfer is described from a glow peak to lower as well as to higher temperature glow peaks. This property has not been described in earlier books. This chapter also describes the effect of sample temperature on the transfer efficiency for incident light. This phenomenon is used to find the energy levels of the donor trap. Using the monochromatic incident light for photo-transfer, optical trap depth is found for the donor trap.* Studies describing the dependence of transfer efficiency on sample temperature as well as studies with monochromatic light exposure for photo-transfer have possibly not been given in earlier books of this field. *These studies are used to propose a model for photo-transfer in mineral CaF_2 .* Finally this chapter deals with photo-bleaching, which is an important subject in sediment dating. It is shown that in a multi-peak

glow curve photo-bleaching and photo-transfer run together. On prolonged light exposure, all glow peaks, donors, as well as accepters attain a low and nearly stable level. At this stage most of the traps are empty and correspondingly the population of the recombination centers also is negligibly small. *As a result the available option for the photo-excited electrons from any of the traps is simply to get retrapped into any of the vacant traps. In other words a to and fro transfer between the traps continues almost indefinitely. This leads to a nearly unbleachable TL signal which is used as the zero level in the TL dating of the sediments.*

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