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C. M. Sunta

# Unraveling Thermoluminescence

 Springer

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C. M. Sunta

# Unraveling Thermoluminescence

 Springer

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Radiation Protection  
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अपूर्वः कोपि कोशोऽयं  
विधते तव भारती ।  
त्ययतो वृद्धिम् आयाति  
क्षयम् आयाति  
संचयात् ॥

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O, Saraswati ! Goddess of wisdom and learning,  
How unique your treasure house of knowledge is.  
It reduces if you hoard it, and  
becomes richer when you share it !

# Foreword

During the past few decades, thermoluminescence (TL) and related techniques have provided substantive inputs in the field of radiation dosimetry. Today most of the radiation dosimetry applications, be it the environmental, personnel dosimetry, space radiation, geo- and archeo-chronology to the estimation of dose delivered to a patient under radiation therapy are based on TLDs and OSL detectors, with TL offering a distinct advantage of its ability to isolate signals/traps that appear at different temperatures/energies. The traffic of charges between the traps has always been a deeply contested issue and there has been no consensus on any mechanism that can adequately describe the underlying kinetic processes and a physical description of these.

This book by Dr. Sunta presents a state-of-the-art account of the current understanding of the theoretical aspects of TL and presents a lucid account of the basics and includes several new aspects of interpretations that are not found in text books so far. The book assumes a basic knowledge of Physics and Mathematics at an undergraduate level, but then takes the reader to the results at the cutting edge of understanding of the subject in an easy to understand manner. The book begins by presenting an introduction to the basic TL phenomena and the point defects in inorganic solids that may serve as traps and recombination centers. It then describes various models that describe kinetics and shows how inadequate the understanding has been in the past. It then takes the next step to explain methods for the kinetic analysis of TL glow peaks and not only discusses the conventional methods but also presents some new possibilities. It then deals with the concept of quasi-equilibrium and discusses various related issues and presents several dimensions of quasi-equilibrium approximation. As the next logical step it then discusses the growth of TL intensity with radiation dose and here highlights the role of thermally disconnected traps—a concept used in the past, but never given its due. The author derives the supralinearity factors and pre-dose sensitization factors from first principles, and presents a new factor called the sensitization factor to provide a unified explanation to two seemingly diverse aspects of supralinearity and pre-dose effect. The next chapter discusses Photo transferred TL in all possible scenarios and with case studies including its various applications.

I found that this book presents the distilled wisdom of the author Dr. C. M. Sunta, who has a nearly five-decade long experience as a  $24 \times 7$  TL practitioner and as a

pioneer in the conceptual development of the subject. I have myself treated Dr. Sunta as one of my teachers in Luminescence dosimetry and I feel happy that such a book will soon be available to students, teachers, and practitioners and will serve as a useful handbook on concepts and formalism. I feel that this will be a must-read for any one venturing into TL and its application and I am sure this will enrich their science manifold. I compliment and thank Dr. Sunta for his assiduous efforts to present his experience to the younger fraternity.

Ahmedabad, India

A. K. Singhvi



# 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  $\alpha$  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  $\alpha$  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  $s/\beta$ . 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  $\sigma_i$  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.*

Mumbai

C. M. Sunta

# Acknowledgments

I owe my deepest gratitude to my late teacher Dr. A. K. Ganguly who assigned to me the work of developing a thermoluminescence (TL) dosimetry system way back around 1965 for use in the nuclear industry in India.

A major part of this book, which supplements earlier books, is the result of the work I did at the University of Sao Paulo (USP), Brazil. During 1992–1994, my visit to USP was arranged by Emico Okuno. I am thankful to her and to Elizabeth Yoshimura for the work we did jointly at USP. This work appears in Chap. 6 of this book. The collaboration at USP was continued with Prof. Shiguelo Watanabe. I would remain ever thankful to Prof. Watanabe, who in addition to giving academic support was extremely hospitable. He and his wife Dona Renate looked after every need and comfort for me and my wife during our visits to Brazil till mid 2004. I owe special thanks to J. F. D. Chubaci and his wife Rosinha who provided every help in and outside the institute in Brazil. I am thankful to my co-workers, J. F. D. Chubaci, Walter Ayta, Tom Piter, and late R. N. Kulkarni, who contributed immensely to the work we did at USP, part of which appears in Chaps. 3, 4 and 5 of this book.

Though this book was conceptualized and written after my retirement I am thankful to B. C. Bhatt, M. S. Kulkarni, Munish Kumar, Anuj Soni, and N. S. Rawat from BARC who have given substantial assistance in preparing the manuscript. It is no small favor to read a friend's manuscript. B. C. Bhatt and Vinita Puri have been gracious enough to read the manuscript and give valuable suggestions and inputs. However, I bear the responsibility for any mistakes or deficiencies either in substance or language that might have remained unattended. My deep apologies to all those whose names I might have missed to acknowledge.

My boundless thanks to my wife Kamla for her patience, encouragement, and sound advice during the course of writing this book. She stayed the course with me and has often had to realign some of her own priorities in the interests of this book. I thank my children, their spouses, and my grandchildren who have been unstinting in their support and love.

Mumbai

C. M. Sunta

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## About the Author

**Dr. C. M. Sunta** was awarded the M.Sc. degree in Physics from the University of Agra before joining the Bhabha Atomic Research Center. He was awarded the Master of Science degree from University of Rochester, New York during his stint as a Health Physics trainee at Rochester and Brookhaven National Laboratory, New York. He was awarded the Ph.D. in Physics during his research career at Bhabha Atomic Research Centre (BARC), Mumbai. During 1968–1975, he worked on two major Research projects, one under World Health Organization (WHO) and the other under the International Atomic Energy Agency (IAEA) Vienna. The former dealt with exhaustive measurement of background radiations along the coastal areas of Kerala State of India which have radioactivity due to thorium bearing sand and its possible health effects. The IAEA project dealt with the development of radiation monitoring systems using the thermoluminescence (TL) phenomenon. Dr. Sunta continued his basic and applied research on TL simultaneously and heading the research group on this subject. From 1973 to 1975, he worked as an International Atomic Energy Agency's expert in Brazil, helping researchers there develop TL dosimetry systems and enhance applied research in this area. He has 80 peer reviewed papers in journals and about 110 conference presentations and research reports to his credit. In 1990, he was appointed as Director, Radiation Safety Division in the Atomic Energy Regulatory Board (AERB), Govt. of India, from where he superannuated in 1992. Subsequently, he accepted a research assignment in the department of Nuclear Physics at the University of Sao-Paulo Brazil and continued as a visiting Professor till 2004. He continues to pursue his research interest interacting with young researchers of BARC. He is a Fellow of the Luminescence Society of India and a Life Member of the Indian Nuclear Society, the Indian Association for Radiation Protection and the Radiation Biology Association of India.

# Chapter 1

## Introduction: Thermoluminescence and Other Forms of Luminescence

**Abstract** Historical development of the subject of thermoluminescence (TL) is given since its discovery by Robert Boyle in 1663. Highlights of early period include the use of TL detectors in early rocket flights, which lead to the discovery X-rays in Solar spectrum. A brief overview of various types of luminescence phenomena such as photoluminescence, radioluminescence, chemiluminescence, bioluminescence, etc., also is given.

### 1.1 Thermoluminescence: Definition

The term “thermoluminescence” (TL) consists of two words: thermo, meaning heat and luminescence, meaning emission of light. These words may appear to mean that the emission of luminescence is caused by heat. This however is not totally true, since heat has only a secondary role in this case. When heat is the primary cause, the emission is called incandescence, for example, the light coming from a hot filament lamp or a burning charcoal. The primary agents for the induction of TL in a material are the ionizing radiations, namely the X-rays, or radiations from radioactive elements to which the sample is pre-exposed. In some materials, ultraviolet light also may excite TL. When the sample is subsequently warmed up, luminescent light is emitted. The act of warming up stimulates the release of the stored energy in the sample due to pre-exposure to the ionizing radiations. The release of this stored energy produces the luminescence. Therefore, some authors have appropriately used an expanded expression for this emission calling it *radiation induced thermally stimulated luminescence*. Some others have abbreviated it to *radiothermoluminescence* to signify that the TL is induced by the radiations. Some authors call it *thermally stimulated luminescence* (TSL). Presently however, most workers of this field have become accustomed to the abbreviated term TL. Incandescence appears in the visible wavelengths only when the temperature of the heated object goes above about 500 °C when a faint red glow begins to appear. As the temperature is raised further, the object becomes

brighter and brighter. The temperature of the tungsten filament in a light bulb may go up to about 2,500 °C. In contrast, TL may appear at any temperature. It appears in the form of a transient glow, when the sample is heated for the first time after exposure to the ionizing radiations. Repeat heating does not produce any emission, unless the sample is irradiated by Ionizing radiation once again. If the sample is irradiated in ice or at liquid nitrogen or liquid helium temperature, the emission of glow may start at any temperature above the temperature of irradiation. Bhasin [1] has made an instrument to measure a glow curve from LNT to about 350 °C. Using this instrument one may see in LiF:Mg, Ti, a strong glow appearing at about  $-128$  °C (Fig. 1.1). The emission of glow may appear in one or more flashes when the sample temperature is raised slowly. The record of light intensity (measured by an instrument such as photomultiplier tube) against time or against temperature is called a glow curve. Usually, however, the intensity is recorded against the temperature. The individual flashes of light in the glow curve record are called glow peaks. In some samples, for example in blue–green variety of fluorite mineral the flashes (glow peaks) can be visually seen coming one after another if the sample is heated in a dark room after a high dose of X or gamma radiations. In some samples even the color of light differs from one flash to other. The color or the spectral property of the glow peak tells about the TL light emitter species, which may be present as an impurity in the bulk sample.

## 1.2 Early History

The phenomenon of TL was discovered by Sir Robert Boyle [2] who reported to the Royal Society of London on October 28, 1663, the observation of a “glimmering light” when he held a diamond for “a good while upon the warm part of his naked body in the darkness of his bed room”. He repeated the observation by using heat from other sources such as a candle, a heated metal and friction. The phenomenon did not find a proper interpretation for about three quarters of a century. The first clear understanding of this observation was provided by Du Fay [3], who showed that this kind of emission was in fact a delayed phosphorescence. Henri Becquerel [4, 5] detected the occurrence of this phenomenon in certain phosphorescent samples toward the end of nineteenth century. He recognized the ability of certain minerals to retain the effect of irradiation from radioactive substances for a long period. Wiedemann and Schmidt [6, 7] used this phenomenon for the first time to detect the cathode rays and UV light. They irradiated a large number of minerals and inorganic compounds with cathode rays and saw intense emission with long storage life in Manganese-doped  $\text{CaF}_2$  and in mineral fluorite. These two substances are used for the dosimetry of ionizing radiations even today. The work of Marie Curie [8] on induction of TL in the fluorite mineral by exposure to radium roused great interest in this field. The discovery of X-rays gave further stimulus to the work on TL. During 1925–1928, Wick and co-workers