Thermoluminescence (TL) kinetics of CaSO₄ and LiF standard materials: a comparative study

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Abstract
In this paper we present comparative reports on CaSO4 and LiF standard materials. $CaSO_4$: Mn has a very high sensitivity but fading of the dosimetry peaks is relatively rapid. The $CaSO_4$: Tm and $CaSO_4$: Dy each has a stable glow peak at about 220°C. LiF has an effective Z of 8.2 (of. 7.4 for tissue) and the energy response relative to tissue is good. Both the materials are available commercially.

Keywords: TL kinetics, TLD, $CaSO_4$, LiF

INTRODUCTION

The TL model was first presented by Randall and Wilkins (1945) [1]. Suppose that after irradiation, n electrons occupy traps and likewise n holes are in hole traps. Upon heating the material, the electrons are released from traps. It is assumed that the probability for recapture by traps is small, and that the lifetime in the conduction band is short so that the un-trapped electrons are almost immediately swept into luminescence centers with the release of light. A simple model of the TL process is illustrated in Fig.1.

At temperature T the probability of release per unit time is given by

$$\lambda = s \exp(-E / kT)$$

where E is the depth of the energy level for the trap below the bottom of the conduction band, k is the Boltzmann constant and s, which has the dimensions of time $^{-1}$ is known as the pre-exponential or frequency factor. If there are n trapped electrons, then the number, dn, released in time dt at temperature T is

$$-dn = ns \exp(-E / kT) dt \qquad (1)$$

where the minus sign indicates that the number of trapped electrons is decreased. If the sample is heated up so that the temperature rises at a linear rate B so that dT = Bdt, we can now substitute for dt and separate the variables to obtain:

$$dn / n = -(s/B) \exp(-E / kT) dT. \qquad (2)$$

At the start of the heating when T = To let the number of trapped electrons be equal to n_o. When a temperature T has been reached there are n electrons left in traps. Equation (2) can be integrated between these limits to give

$$n = n_o \exp[-\int (s/B) \exp(-E / kT) dT]. \qquad (3)$$

The intensity, I, of the thermoluminescence is proportional to the rate of filling of luminescence centers which in turn is equal, on this model, to the rate of emptying of traps, i.e. $I \propto dm/dt = -dn / dt$ (m is the density of luminescence centers) so that, from equation (1)

$$I = A n s \exp(-E / kT) \qquad (4a)$$

where A is the constant of proportionality. If we substitute for n from equation (3) then we have

$$I = A n_o s \exp(-E / kT) \exp[-\int (s/B) \exp(-E / kT) dT]. \qquad (4b)$$

A plot of I against T is known as a glow curve. The rate of emptying of traps on this model depends on the first power of the number of trapped electrons and such kinetics are known as *first order or mono-molecular* kinetics.

Garlick and Gibson (1948) [6] considered the case where electrons may be re-trapped in addition to undergoing re-combinations with holes. The differential equation obtained from this model has the form

$$-dn / dt = (n^2 s / N) \exp(-E / kT) \qquad (5)$$

where N is the total number of electron traps. Here n is raised to the power two and we thus have *second-order or bimolecular* kinetics.

First- and second-order glow curves differ in a number of respects. First-order glow curve rises steadily to the peak and then falls rapidly to zero as the temperature rises, the second-order curve is more nearly symmetrical. Also, whilst the peak temperature in the first-order case is independent of the initial number of trapped electrons (and hence, in practical terms, of the radiation dose), the second-order curve shifts to progressively lower temperatures as the number of trapped electrons is increased. This arises because, in the second-order case, the fractional rate of electron release, $-(1/n).(dn/dt)$ is in itself a function of n (see equation 5).

More complex situations are possible. Moreover, real solids will usually have a number of trapping levels (and perhaps several types of luminescent center) and will exhibit a glow curve having several peaks. Some applications have been discussed below-

In the last three decades extensive studies have been

undertaken on thermoluminescence(TL) and TL related phenomena, new and sensitive TL phosphors were developed. TL is a very good technique used in research in basic as well as applied sciences. It is already applied in various fields and is now extended to a whole spectrum of disciplines such as archaeology, forensic sciences, geology, radiation dosimetry, radiation physics, solid state physics, space sciences and many more[7-9] . Some other luminescence techniques are also finding useful applications.

EXPERIMENTAL

All the materials were synthesized by solid state diffusion method.

RESULTS AND DISCUSSION
TL dosimetry in Calcium sulphate CaSO₄

$CaSO_4$:Mn has a very high sensitivity but fading of the dosimetry peaks is relatively rapid. $CaSO_4$:Tm and $CaSO_4$: Dy each have a stable glow peak at about 220°C. All Ca bearing phosphors have a high effective atomic number and so over-respond to low-energy photons. TLDs are now finding increasing use in personnel dosimetry where they are tending to replace the film badge. It is in this area that automated TLD systems are of particular importance.

A TL dosimeter will normally have several elements. A phosphor covered by ~ 5 -10mg cm $^{-2}$ of tissue-equivalent absorber allows the dose to the basal layer cells of the skin to be determined; whereas an element covered by ~ 400-1000 mg cm $^{-2}$ allows the deep dose to be measured. Where neutrons are present a neutron-sensitive element can be included. For example a phosphor containing mainly ^{6}Li will respond to neutrons and gamma rays whereas one containing ^{7}Li responds only to gamma rays.
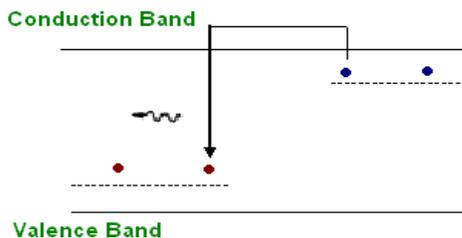

Fig 1. A simple model of the thermoluminescence process. Upon heating the material electrons may be released from traps to the conduction band. From there they can recombine with trapped holes with the emission of light.

TL dosimetry in Lithium Fluoride LiF:

Lithium fluoride doped with Mg and Ti is probably the most widely used TL dosimetry material. The details of the TL process in LiF:Mg,Ti are still not fully established but it seems that defect complexes consisting of combinations of Mg $^{2+}$ and Li $^+$-vacancies act as trapping centers. Ti ions are probably associated with the recombination centers. Suitably doped LiF is available commercially as single crystals, powders or powders held in a PTFE binder. LiF in which the ^{6}Li isotope is enriched is available for use in neutron dosimetry.

LiF has an effective Z of 8.2 (of. 7.4 for tissue) and the energy response relative to tissue is good. After readout, care must be taken to apply a suitable anneal cycle in order to remove any residual TL and to restore the phosphor sensitivity.

In addition to personnel dosimetry, TLDs can be used to determine doses absorbed during radio-therapy and diagnostic radiology. (see for example, McKinlay, 1981[2] ; Becker, 1973 [3]; Oberhofer and Scharmann, 1981[4]; McKeever, 1984[5]; Horowitz, 1984[6]; Stoebe, 1984 [7]

CONCLUSIONS

In the last three decades extensive studies on CaSO4 and LiF standard materials have been undertaken on thermoluminescence (TL) and TL related phenomena. They are very good phosphors used in research in basic as well as applied sciences. It is already applied in various fields and is now extended to a whole spectrum of disciplines such as archaeology, forensic sciences, geology, radiation dosimetry, radiation physics, solid state physics, space sciences and many more.

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