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ARAB REPUBLIC OF EGYPT

ATOMIC ENERGY ESTABLISHMENT

RADIATION PROTECTION DEPARTMENT

TRAPPING PARAMETERS OF NATURALLY OCCURING THERMOLUMINESCENT MATERIALS

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ABSTRACT

The trapping parameters, namely, the frequency factor and activation energy for natural quartz, feldspar and barite are investigated. The frequency factor is calculated using the Randell and Wilkins model whereas the initial rise method is viewed suitable to evaluate the trap depth. The inconsistency between the experimentally and theoretically glow curves are attributed to the retrapping effects and randum impurity levels leading to numerous overlapping peaks.

INTRODUCTION

The term thermoluminescence (TL) refers to re-emission of fraction absorbed energy in the form of visible light upon heating the substance. Also, the glow curve being the temperature dependence of the light output.

Indeed during the last decade, thermoluminescence starts to be the most developed tools for radiation dosimetry mostly using commercial materials.[1] Consequently it is recommended to be used with fair accurecy, in personnel monitoring, medical Physics as well as dosimetric measurements.[2]&[3] Recently, the natural materials pertinent provide a very good tool as thermoluminescent dosimeters as they are quite available in large quantities and cheap. The availability of which overcomes the problem of reusability that causes conflecting results[4]. The main task of this work is to throw some light on the TL mechanism of some naturally occuring material with special regard to the trapping parameters.

The kinetic study of TL mechanism are generally simplified in a way first introduced by Randell and Wilkens[5]. Accordingly, assuming a linear heating rate, the emitted light intensity for the first order processes is given by

$$I\ (T) = Cn_o S \exp \left[\int_{T_o}^{T} S/q\ e^{-E/kt}\ dT \right] \exp\ (-E/kT) \qquad (1)$$

and

$$I(T) = Cn_oS\, e^{-E/kT} \Big/ \left[1 + S/q \int_{T_o}^{T} e^{-E/KT}\, dT\right]^2 \qquad (2)$$

For the second order process, where C is the constant of proportionality, n_o is the number of electrons (holes) at $T = 0$, K is Boltzmann's constant, E is the activation energy,
S is the frequency factor (Sec^{-1}), T is the absolute temperature and q is the heating rate.

Provided a maximum glow,, then
$dI/dT = 0$, which gives

$$qE/SK\, T_m^2 .\ \exp\left(-\frac{E}{K T_m}\right) = 1$$

For the first order , and

$$\frac{q E}{SK\, T_m^2} \exp\left(E/kT_m\right) = 1 + \frac{2K\, Tm}{E} \qquad (3)$$

For the second order in which T_m is the peak temperature.
Equation (1) can be proximated to

$$I = Cn_o S \exp - \left[\left\{(ST/q).\ F(E/KT) + (E/KT)\right\}\right] \qquad (4)$$

where $F(E/KT) = \exp.(-E/KT) / (E/KT)$
provided $E/KT > 25$

The use of this approximation facilitate its evaluation at various T and the theoretical glow Curve can thus be Calculated.[7]

EXPERIMENTAL

The naturally occuring TL materials under investigation are feldspar, quartz and barite. The samples being cut, washedthoroughly and annealed at 400 ^{o}C for one hour to remove the TL background signals. The samples then irradiated by gamma rays up to $1.1x10^{4}$ rad using Co^{60} – γ - ray unit.

The glow curves are measured using Harshaw 2000 (A+B) read out unit which is connected to X-Y recorder.

The heating rate is $10.5^{o}C. S^{-1}$ and the maximum tray temperature is 350 ^{o}C reached in 30 Seconds.

For trap depth determination the initial rise method as suggested by Giarlik and Gribson is used. (8) In which the activation energy (E) is related to the light intensity (I) in the form.

$$\ln I = \text{Constant} - E/KT$$

The plot of $\ln (I)$ Versus T^{-1} for temperature well below T_m is straight line with the slope E/K.

RESULTS AND CALCULATION

The experimental glow curve for feldspar and the relation between the logarithm of light intensity versus 1/T as well as the theoretically calculated glow curve are shown in Fig (1). It is clear that the glow curve assumes a single peak with maximum glow at 393 K (Curve a). The straight line behaviour of the first rising portion of curve (b) insures that initial rise method is quite proper for the determination of E particullarly at low light intensities, since as the temperature increases towards the peak value a remarkable deviation from the exponential is noticed. Similarly for quartz the above mentioned trends are observed, Fig (2)

However, in case of barite the dosimetric peak at 423 K is disturbed by a neighbouring peak at 393 K. The latter one is removed by the cleaning technique as suggested by Hoogenstreaten[9] i.e. quick heating of the sample up to 80°C in 10 seconds Fig.(3)

Since each glow peak is associated with electron traps of single energy depth, it is possible to estimate the trap activation energy (E) using the slopes of figures (1÷3) and the frequency factor (S) Can be evaluated using equation (3) and (4) for first and second order process respectively. Table (1) summerises the experimentally evaluated (E) and the calculated frequency factor (S) for the materials under investigation.

Table (1)
Value of Trap Depth and Frequency Factor for Naturally Occuring TLD Materials

TL Material	Dosimetric Peak K	Trap Depth eV	Frequency Factor First order s^{-1}	Frequency Factor Second order s^{-1}
Feldspar	393	0.83	3.04×10^{10}	2.81×10^{10}
Quartz	393	0.81	1.6×10^{10}	1.48×10^{10}
Barite	423	0.91	4.5×10^{10}	4.17×10^{10}

Provided, the activation energies experimentally evaluated the corresponding theoretical glow curves are calculated as shown in Fig (1c - 3c). In general there is consisting between experimental and calculated glow curves. However in particular, the ascending part of the theoreticlly glow curve is in good accordance with the experimental one in case of feldspar, whereas, the descending part experiences a sharp decrease.

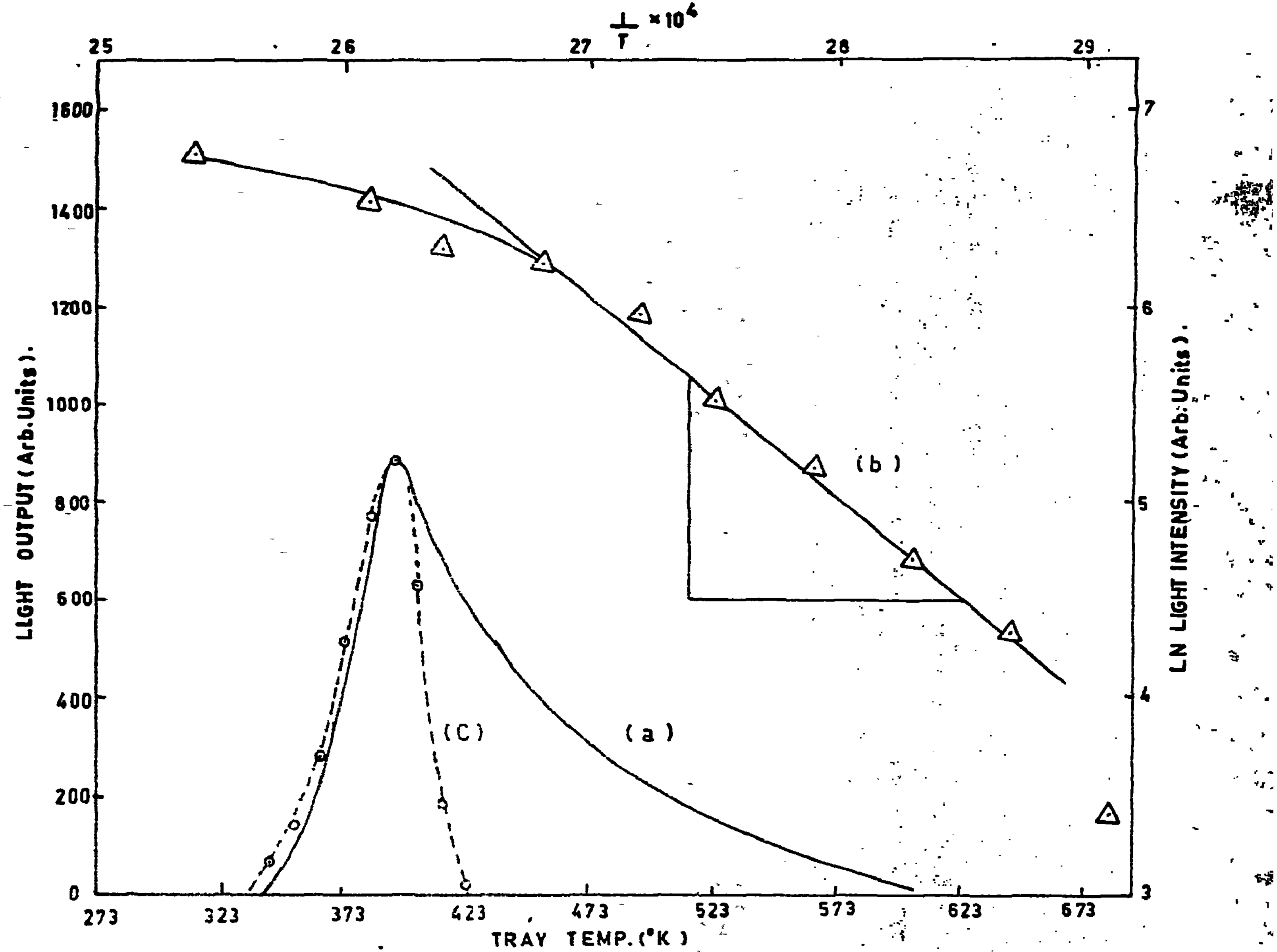

FIG.(1) LIGHT INTENSITY DEPENDENCE ON TRAY TEMPERATURE FOR FELDSPAR.
a) EXPERIMENTAL GLOW CURVE, b) LN LIGHT INTENSITY V_s T^{-1} & c) CALCULATED GLOW CURVE.

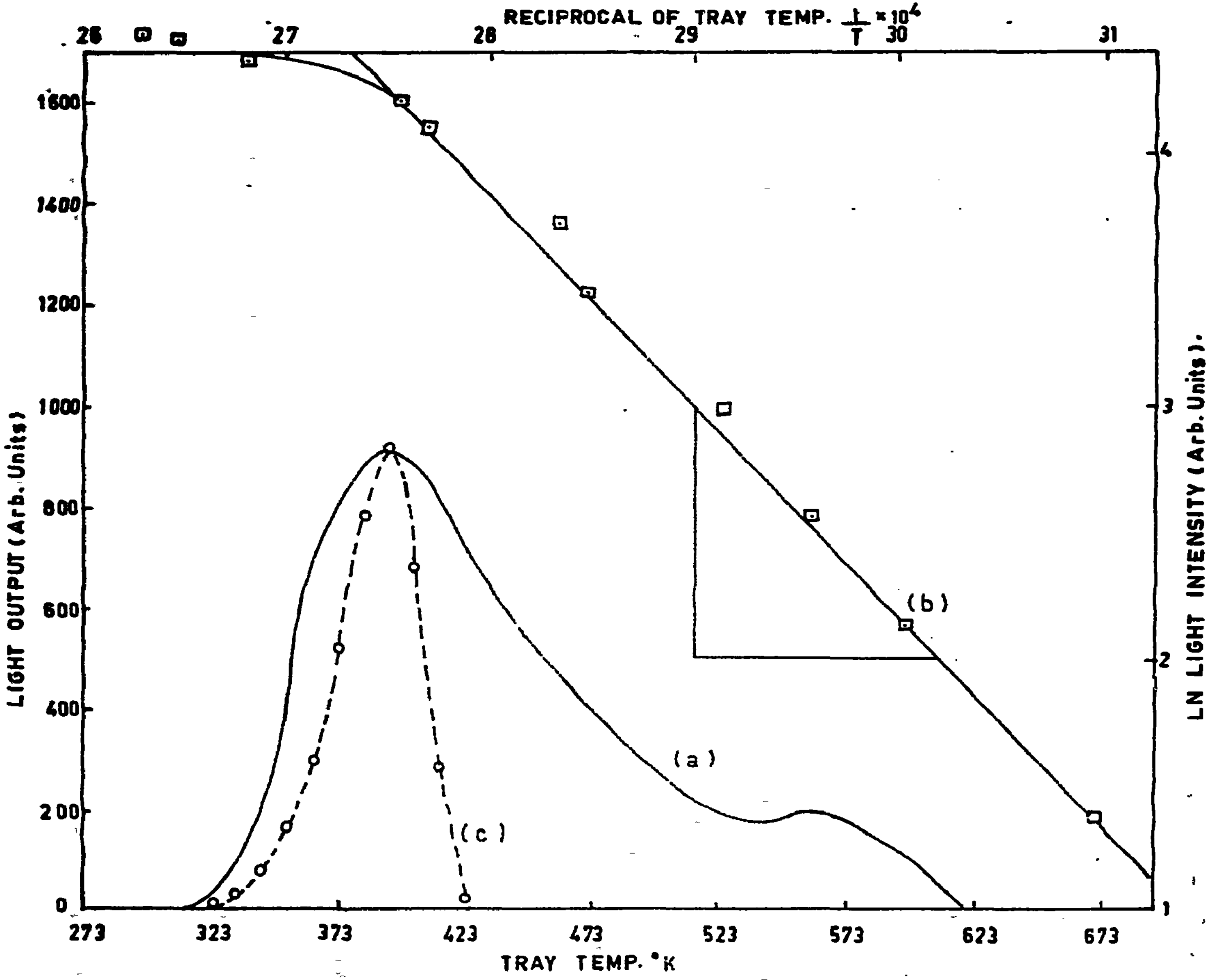

FIG.(2) LIGHT INTENSITY DEPENDENCE ON TRAY TEMPERATURE FOR QUARTZ.
a) EXPERIMENTAL GLOW CURVE, b) LN LIGHT INTENSITY Vs T^{-1} & c) CALCULATED GLOW CURVE.

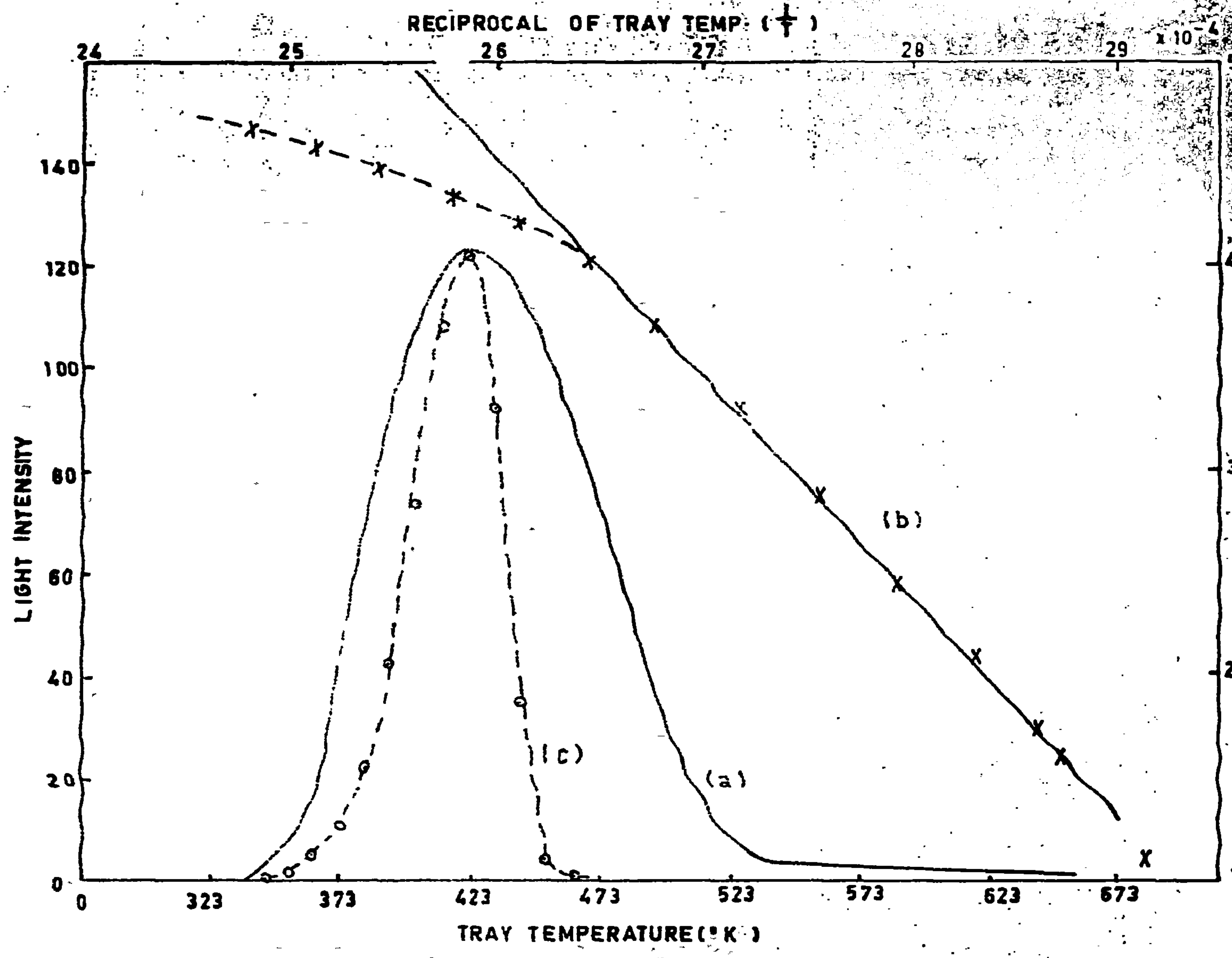

FIG.(3) LIGHT INTENSITY DEPENDENCE ON TRAY TEMPERATURE FOR BARITE
a) EXPERIMENTAL GLOW CURVE ; b) LN LIGHT INTENSITY Vs T^{-1} & c) CALCULATED GLOW CURVE.

DISCUSSION AND CONCLUSION

Although it is clear that the glow curve deviates from the exponential behaviour at the relatively higher temperatures but the initial rise method for activation energy determination being quite proper. That is and it has frequently been assumed that the second order process does not contribute too much for thermoluminescent mechanism which appears untrue since its frequency factor (Tab. 1) is comparable with that for the first order further discussion will be followed.

Provided $E \simeq 25\ KT_m$,, the approximation method is applicable(10) which give rise to the same values of activation energy as that obtained by the initial rise method.

The general pattern adopted in this subject is that the discribancy between theory and experiment is attributed to the retrapping effect which can be enhanced even for the occuring materials under investigation. This is because no significant change in the glow curve is noticed regarding the previous work on thermoluminescence dosimeters.

Ultimately, all models discussed so far consider only single set of impurity level at certain descrete energy. However a real naturally occuring crystals contains in many cases a range of trapping centres which leads to a hole spectrum of glow peaks consisting oftenly numerous overlapping peaks.

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