Numerical solutions of Schön-Klasens model for luminescence efficiency

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Abstract. Phosphors exhibit luminescence following irradiation and the absorption of energy depends upon the relative probabilities of the radiative and non-radiative transitions. In general, the luminescence efficiency of a phosphor is related to the probability of a luminescence transitions and probability of a non-radiative transitions. Several experimental measurements of the luminescence efficiency have shown it to be strong temperature dependent over given temperature ranges. In literature, Schön-Klasens model has been offered to explain the temperature dependence of luminescence efficiency. In this study, theoretical basis and some numerical solutions of the model was discussed. The brief information about mathematical principles of the model was given and differential equations expressing the charge carrier transitions were derived. Numerical solutions of the equations were performed by using Mathematica 8.0 computer code. All the analysed parameters were chosen realistically. According to the simulations, glow curves intensities were decreased by the appropriate η_e and η_h ratio.

1 Introduction

A number of models are available in the literature for the explanation of the thermoluminescence (TL) event. The theoretical explanation of TL is based on the electron band theory of an insulating or semiconducting solid. It consists of a set of localized energy levels in the forbidden band, which arises due the presence of impurities and other point defects. The energy levels act as traps and recombination centres in the TL process [1-5]. All TL phenomena are governed by the process of the electron hole recombination. It should be noted that rather complex processes are taking place in the traffic of charge carrier between trapping states and luminescent recombination centres. Almost all of TL models have been based on the consideration of charge release from electron trap only. In this paper, we assumed that not only electrons but also holes are mobile in the same temperature interval. The model introduced originally by Schön and colleagues [6,7] and used by Klasens [8]. According to the model holes also contribute to TL emitting like electrons. Fig. 1 show that energy levels, charge carrier transitions and related parameters suggested by the Schön-Klasens model [4].

According to the model charge carrier traffic are given in Eq.1-4. The 4 equations set also describe the simultaneous release of holes during the thermal stimulation of the trapped electrons [1-4].

$$\frac{dn}{dt} = -s_e n \exp(-E_e/kT) + A_{te} n_c (N-n) - A_{rh} n_v n \quad (1)$$

$$\frac{dn}{dt} = s_e n \exp(-E_e/kT) - A_{te} n_c (N-n) - A_{re} n_e m \quad (2)$$

$$\frac{dm}{dt} = -s_h m \exp(-E_h/kT) + A_{th} n_v (M-m) - A_{re} n_e m \quad (3)$$

$$\frac{dn_v}{dt} = s_h m \exp(-E_h/kT) - A_{th} n_v (M-m) - A_{rh} n_v n \quad (4)$$

Figure 1. Generalized energy levels scheme and allowed transitions for Schön - Klasens model [4].

In here, the instantaneous concentration of electrons in the conduction band is denoted by n_e (m^{-3}) and that of holes in the valance band by n_v (m^{-3}) respectively. N (m^{-3}) denotes here the total concentration of electron trapping states which is a constant and n (m^{-3}) the instantaneous concentration of filled electrons trap which is a variable. E_e (eV) and s_e (s^{-1}) are the activation energy and frequency factor of the electron trap, respectively, k is the Boltzmann constant ($eV K^{-1}$) and A_{te} ($m^3 s^{-1}$) is the trapping (re-trapping during heating) probability of electrons from the conduction band. M (m^{-3}) denotes here the total concentration of hole trapping states which is a constant and m (m^{-3}) the instantaneous concentration of filled holes trap which is a variable. E_h (eV) and s_h

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(s^{-1}) are the activation energy and frequency factor of the hole trap, respectively. A_{th} ($m^3 s^{-1}$) is the probability of capturing hole in M, whereas A_{re} ($m^3 s^{-1}$) is the recombination probability of free electrons with captured holes. A_{rh} ($m^3 s^{-1}$) is the recombination probability of free holes with captured electrons in electron trap.

At the same time, the equations to keep to the right neutralization condition expressed in Eq 5.

$$dn/dt + dn_c/dt = dm/dt + dn_v/dt \quad (5)$$

If all recombination events are radiative and produce photons and all photons are detected, TL glow curve is expressed by Eq.6. [1-4].

$$I_{TL} = \eta_e I_{TLn} + \eta_h I_{TLh} \quad (6)$$

For the equations set approximate solutions were given by Bräunlich and Scharmann [9]. These authors considered four extreme cases, involving the rates of electron and hole retrapping and their comparison with the corresponding recombination rates. The model also solved numerically by McKeever et.al. [10] without any of the assumptions of the Bräunlich and Scharmann and reached the same conclusions. In addition to these the model was solved numerically by Uzun [11] for various trapping and recombination probabilities.

2 Methodology

In this study, we assumed that material irradiated before heating stage and has electrons in electron trap (n_o) and holes in hole trap (m_o). In this case it is important assumption that there are not any charge carriers in the conduction and valence bands. This is followed by a heating stage. During this stage M and N are assumed to be rather far from the valence band and conduction band, respectively. Electrons from N may be thermally released into the conduction band and then either re-trap in N or recombine with holes in M. At the same time holes from M may be thermally released into the valence band and then either re-trap in M or recombine with electrons in N. For a given set of trapping parameters, differential equations governing the process during the excitation stage were numerically solved by using a special code in the Mathematica 8.0 [12] computer program with an explicit Runge-Kutta method [13]. During the solutions temperature was changing with a constant heating rate (β) and therefore instantaneous temperature is expressed by Eq. 7.

$$T = T_o + \beta t \quad (7)$$

Where T_o is the initial temperature at the beginning of heating stage and t is the time (s). Both recombination into N and M are considered to be radiative, but separable. Thus, the intensity in photons per m^3 per second of one spectral component of TL is proportional to the rate of change of N, i.e. Eq. 1 and the second spectral component is assumed to be proportional to the rate of change of M, namely, Eq. 3. The shape, position and intensity of the glow curve are related to a various

trapping parameters of the trapping states responsible for the TL emission.

3 Results

Thermoluminescence intensities come from only electrons and only holes movement were determined and glow curve was calculated. Results are given in Fig. 2 and parameters used in the simulations were given in Table 1. It can be seen from the Fig. 2 that contributions of the electrons and holes on glow curve are the same for the given trap parameters in Table 1.

Figure 2. Thermoluminescence intensity comes from only electrons (black line), holes (red line) and glow curve (green line)

Table 1. Trap parameters for Fig.2

Parameters	Values
$E_e = E_h$ (eV)	1.00
$s_e = s_h$ (s^{-1})	10^{12}
$M = N$ (cm^{-3})	10^{10}
$m = n$ (cm^{-3})	10^{10}
$A_{te} = A_{th}$	10^{-9}
$A_{re} = A_{rh}$	10^{-7}
$\eta_e = \eta_h$	1.00

Figure 3. Contributions of the electrons on glow curve for different η_h values.

The effect of the radiative recombination ratios of the electrons and holes on glow curves are shown in Fig.3-5. Trap parameters used in the simulations were given in Table 2.

Figure 4. Contributions of the holes on glow curve for different η_h values.

Figure 5. Glow curves for different η_h values.

Table 2. Trap parameters for Fig.3-5.

Parameters	Values
η_e	1
η_h	1.00-0

The effect of the radiative recombination ratios of the electrons and holes on maximum thermoluminescence intensities of the glow curves are shown in Fig.6.

4 Conclusions

In this study Schön-Klasens model has been solved by numerically. In the solutions η_e and η_h parameters were chosen as variables and others were constant. By using these parameters, Eq.1 to Eq.6 are solved by numerically but no simplifying assumptions had been made.

Simulations show that according to Schön-Klasens model, in which electron and hole can be released by thermally, thermoluminescence glow curve is shaped by charge carrier movement resulting recombination. This

process is different from the other models' process because, now hole is not a stable charge carrier.

Figure 6. The effect of the η_h on I_M . Maximum intensities of the glow curve, electron and hole contributions are shown as black triangle, red and blue circles, respectively.

According to simulations;

- When $E_e=E_h$ and $\eta_e=\eta_h$, glow curve is shaped by both electrons and holes movement. In addition to this, contributions of them to glow curve are the same ratios.
- When $E_e=E_h$ and $\eta_e>\eta_h$, glow curve shaped by both electrons and holes movement. But in this case contribution of electrons is bigger than holes depending on η_e/η_h ratio.
- When $\eta_h=0$, glow curve is shaped by only electrons.
- When $\eta_e=0$, glow curve is shaped by only holes.
- When $\eta_e=1$, it means that all electron-hole (electron is mobile and hole is still) recombination are radiative and vice versa if $\eta_e=0$.

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