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A quantitative kinetic model for Al$_2$O$_3$:C: TL response to UV-illumination

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Abstract

This paper presents a quantitative kinetic model applicable to the important dosimetric material Al$_2$O$_3$:C. The model describes successfully the thermoluminescence (TL) response of the material to UV illumination (206 nm). The energy levels in this model consist of the main dosimetric trap, two competing deep hole and deep electron traps, and the luminescence center. The model also describes successfully the experimental variation of the optical absorption coefficient $K$ with UV fluence. The values of the kinetic parameters are not arbitrary, but are obtained either from published experimental data, or by using reasonable physical assumptions. A correction factor is applied to the calculated UV-fluence to account for the fact that the samples used in the experiments were several optical lengths thick. By using this correction factor, the experimental data can be shown on the same graph as the calculated curves of TL vs. UV-fluence and $K$ vs. UV-fluence, with the UV fluences given in photons/cm$^2$ and not in some arbitrary units.

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1. Introduction/Scope

The study of the thermoluminescence (TL) response of Al$_2$O$_3$:C to ionizing radiation and to UV radiation is of great practical importance in dosimetric applications. There have been several previous attempts to produce theoretical models to describe various aspects of the TL response of this material to ionizing radiation (see for example Agersnap Larsen, 1999; Pagonis et al., 2006, 2007, and references therein).

Recently we have presented a quantitative model (Lawless et al., 2005; Chen et al., 2006; Pagonis et al., 2006, 2007) for the TL and OSL response of Al$_2$O$_3$:C to ionizing radiation. The model provided a mathematical description for several experimental results: the TL vs. beta dose behavior, the variation of the absorption coefficient $K$ with the beta dose and the nonmonotonic behavior of TL and OSL at high doses.

Yukihara et al. (2003) carried out a comprehensive experimental study of the effect of deep traps on the thermoluminescence (TL) of Al$_2$O$_3$:C by using both beta irradiation and UV-illumination. The concentration of F and F$^-$ centers in the samples were monitored by optical absorption measurements, and competing deep holes and deep electron traps were identified.

In this paper it is shown that the same model can also describe the corresponding experiments with UV-illumination instead of beta irradiation. The study of Yukihara et al. (2003) involved three samples from different batches labeled D320, Chip101 and B1040. Their data showed that the maximum height of the TL peak at 450 K increased monotonically as a function of the UV illumination period. The UV data showed no decrease of the TL signal at high UV-fluences, in contrast to the drop in the corresponding TL signal observed at high beta doses. The value of the TL signal reached a saturation value for samples D320 and Chip 101, while for sample B1040 the saturation point was not reached even at the highest value of the UV-fluence used in the experiments.

In a second series of experiments, these authors studied the variation of the absorption coefficient $K$ of the samples with the UV illumination period. The value of $K$ increased monotonically with the UV irradiation time, up to different saturation values for all 3 samples used. The authors noted that sensitization occurred after either beta or UV irradiation, and associated these sensitization phenomena with the filling of the
The set of simultaneous differential equations governing the above process (206 nm) excites electrons from the F-centers to the conduction band according to the process $F + h\nu \rightarrow F^* + e^-$. This process produces free electrons that can be captured by electron traps only, at the same time causing a conversion of the F centers into $F^+$ centers. The present kinetic model consists of the dosimetric trap, competing deep hole and deep electron traps, and of the luminescence process.

Several previous studies of the UV-response of Al$_2$O$_3$:C showed that irradiation with UV (206 nm) excites electrons from the F-centers to the conduction band according to the process $F + h\nu \rightarrow F^* + e^-$. This process produces free electrons that can be captured by electron traps only, at the same time causing a conversion of the F centers into $F^+$ centers. The present kinetic model consists of the dosimetric trap, competing deep hole and deep electron traps, and of the luminescence process.

The set of equations (1) through (6) are valid for both the irradiation and heating stages of the simulation.

The TL intensity $I(T)$ is associated with the electron-hole recombination in the recombination center $m_1$, and is given by

$$I(T) = A_{m_1} m_1 n_1 \eta(T).$$

The temperature-dependent factor $\eta(T)$ describes the thermal quenching of the TL intensity and is given by

$$\eta(T) = \frac{1}{1 + C_1 \exp(-W_j/kT)}.$$
The optical lengths of the sample, these two quantities must be... 

\[ \text{Feff} = \frac{\int_{x=0}^{x=L} e^{-Kx} \, dx}{L} = \frac{F_o e^{-KL}}{L} \]

Equation (9) gives the correction factor of 0.243 by which the nominal UV-fluences must be divided in order to account for the thickness of the samples, in agreement with the results shown in Figures 1 and 2.

Figure 2 shows the variation of the concentration of holes in the luminescence center (\(m_1\)) as a function of the UV-fluence for sample Chip101. The value of \(m_1\) is proportional to the absorption coefficient \(K\) of the material, and can be directly compared with the experimental data given in Figure 9a of Yukihara et al. (2003). The experimental data are also shown as solid circles in Figure 2. The x-axis of the calculated UV-fluence in Figure 2 has also been corrected using the same correction factor of 0.243. The agreement between theory and experiment is seen to be reasonably good, especially considering that only 7 experimental data points are available over more than 2 orders of magnitude of the UV fluence.

Figure 3 shows the detailed behavior of the concentrations of holes and electrons in the model at the end of the relaxation period for sample Chip101. The calculations in Figure 3 show that the dosimetric peak (\(n_1\)) reaches saturation at a UV-fluence of \(\sim 2 \times 10^{15}\) photons cm\(^{-2}\), while the competitor deep electron trap (\(n_2\)) continues to rise with UV-fluence. The \(F^+\) center concentration (which is proportional to \(m_1\)) rises continuously for all fluences shown in Figure 4, while the deep hole traps (\(m_2\)) and the valence band do not participate in the UV-illumination process (\(m_2 = n_v = 0\)). The maximum of the TL peak (\(T_{\text{max}}\)) rises to saturation at about the same UV-fluence as the dosimetric trap \(n_1\).

It must also be noted that the simulated TL glow curves have the expected shape for first order kinetics, but they do not have the experimentally observed glow curve widths. The simulation shows an almost constant peak width of \(\sim 34\) K, while the experimental data of Yukihara et al. (2003) show a widening of the TL glow curve with increasing UV-fluence. Another point of disagreement between theory and experiment is the predicted temperature of the maximum of the TL intensity (\(T_{\text{max}}\)). While theory predicts a constant \(T_{\text{max}} = 453\) K at all UV-fluences, the experiment shows a systematic shift of the \(T_{\text{max}}\) with the UV-fluence, towards higher values of \(T_{\text{max}}\). This disagreement between the experimental and the calculated TL glow curves was also observed during the previous simulation of the TL dose response graphs for this sample (Pagonis et al., 2007).

This discrepancy is easy to understand because previous experimental studies have shown that the dosimetric peak at 450 K has a composite structure. This structure has been interpreted as either involving two components corresponding to the release of both electrons and holes from the traps, or as a series of overlapping first order TL peaks (Agersnap et al., 1999; Colyott et al., 1996; Whitley and McKeever, 2000). This is obviously a feature not included in the current simplified model.

Discussion and Conclusions

The model presented in this paper was used previously to provide a mathematical description of the following...
experiments: TL vs. beta dose behavior, variation of the absorption coefficient $K$ with the beta dose and nonmonotonic behavior of TL and OSL at high doses. This paper showed that the same set of kinetic parameters in the model can also describe the experimental TL vs. UV-fluence behavior of $\text{Al}_2\text{O}_3$:C, as well as the closely related variation of the optical absorption coefficient $K$ with UV-fluence. The values of the kinetic parameters in this model are based on reasonable physical assumptions and on available experimental data. A correction factor was applied to the calculated UV-fluences to account for the fact that the samples are 4 optical lengths thick. The experimental data are shown on the same graph as the calculated curves, with the UV-fluences given in photons/cm$^2$ and not in some arbitrary units, as is often the practice in TL kinetic models.

A more comprehensive model for this material must include: (a) the known shallow TL traps in this material, (b) thermal dissociation effects for the deep hole traps and deep electron traps at appropriate temperatures, and (c) localized transitions between F and F$^-$-centers and (d) the effect of finite thickness of the sample, especially at large doses.

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References


