Available online at www.sciencedirect.com
SCIENCE DIRECT
Nuclear Instruments and Methods in Physics Research A 537 (2005) 443–445
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Afterglow, low-temperature radioluminescence and thermoluminescence of Lu_2O_3:Eu ceramic scintillators

L.A. Kappers[a,*], R.H. Bartram[a], D.S. Hamilton[a], C. Brecher[b], A. Lempicki[b]

[a] Department of Physics, University of Connecticut, U-3046, 2152 Hiilside Road, Storrs, CT 06269-3046, USA
[b] ALEM Associates, 303A Commonwealth Ave., Boston, MA 02115, USA

Available online 27 August 2004

Abstract

Comparison of thermoluminescence (TL) and scintillation light outputs with continuous gamma-ray excitation reveals that the concentration of deep hole traps in ceramic Lu_2O_3:Eu, tentatively attributed to anion Frenkel defects, is enhanced by reversible radiation damage. Shallow electron and hole traps are tentatively attributed to surface states at grain boundaries. Electrons in shallow traps serve as non-radiative recombination centers. A model for anomalously persistent afterglow following pulsed X-ray excitation is based on a continuous distribution of hole traps inferred from TL following extended gamma-ray irradiation at $-135\,^{\circ}C$.
© 2004 Elsevier B.V. All rights reserved.

PACS: 78.60.Kn; 78.60.Ya; 29.40.Mc; 81.05.Je

Keywords: Scintillators; Radioluminescence; Thermoluminescence; Afterglow; Ceramic Lu_2O_3:Eu

1. Introduction

A method based on comparison of thermoluminescence (TL) and scintillation light outputs as functions of radiation time has been devised to investigate traps that limit efficiency of scintillator response by competing with prompt recombination at activators [1–3]. This method is presently applied to transparent ceramic europium-activated lutetium sesquioxide (Lu_2O_3:Eu) [4]. An estimated value of the 4*f* binding energy of Eu^{3+} in Lu_2O_3 with respect to the binding energy of the valence band maximum is $\approx 2.0\,eV$. Consequently, Eu^{4+} is unstable in Lu_2O_3 and electrons are captured first by activators to form Eu^{2+}. Holes subsequently either recombine to form excited activators, $Eu^{3+}*$, or are diverted to hole traps.

2. Experiment

An electron Van de Graaff accelerator with the electron beam stopped by a thin copper target is

*Corresponding author. Tel.: +1-860-486-4782; fax: +1-860-486-3346.
E-mail address: kappers@uconnvm.uconn.edu (L.A. Kappers).

0168-9002/$ - see front matter © 2004 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2004.08.060

employed as a gamma-ray source and a common apparatus and detection scheme is utilized for both TL and scintillation measurements [1]. This method, recently extended to accommodate irradiation at cryogenic temperatures, exploits a common activator with identical emission spectra for both measurements. Light output and sample temperature are plotted as functions of time in Fig. 1 for a 16 min irradiation at low temperature. Similar data were recorded at both low temperature and room temperature for radiation times ranging from 15 s to 16 min.

3. Rate equations

Room-temperature irradiation reveals that the concentration of deep hole traps is enhanced by self-limiting, reversible radiation damage, and that the traps ultimately saturate. Low-temperature irradiation reveals that shallow hole traps exhibit neither radiation enhancement nor saturation, but that radiation induces non-radiative recombination centers attributed to electrons in shallow electron traps. The data recorded at low temperature can be modeled by rate equations with approximate solutions:

$$\tilde{n}^{(\mathrm{se})} \equiv n^{(\mathrm{se})}/f' \cong \frac{\sqrt{1+2\beta t}-1}{\beta} \tag{1a}$$

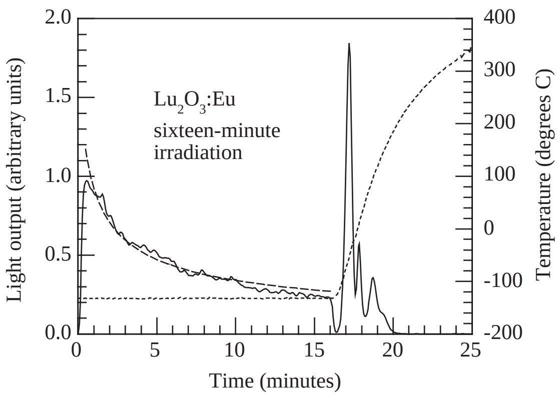

Fig. 1. Light output (continuous line) and sample temperature (dotted line) as functions of time for a Lu_2O_3:Eu ceramic scintillator irradiated at low temperature. Simulation (dashed line) is based on Eqs. (1) and (3) and on Table 1.

$$\beta \equiv \frac{f' A_{\mathrm{NR}}}{n_{\mathrm{e0}} A_{\mathrm{r}} + N^{(\mathrm{sh})} A^{(\mathrm{sh})}} \tag{1b}$$

$$\frac{G(s)}{S} \cong \frac{N^{(\mathrm{sh})} A^{(\mathrm{sh})}}{n_{\mathrm{e0}} A_{\mathrm{r}}} = \text{constant} \tag{2}$$

$$L.O. \propto n_{\mathrm{a}} \tau_{\mathrm{r}}^{-1} \propto \frac{f'}{(1+G(s)/S)(1+\beta \tilde{n}^{(\mathrm{se})})} \tag{3}$$

$$\frac{G(s)+S}{t} \propto \left(\frac{1+G(s)/S}{G(s)/S}\right) \frac{\tilde{n}^{(\mathrm{sh})}}{t} = \frac{\tilde{n}^{(\mathrm{se})}}{t} = \frac{\sqrt{1+2\beta t}-1}{\beta t}, \quad \tilde{n}^{(\mathrm{sh})} \equiv n^{(\mathrm{sh})}/f' \tag{4}$$

where $G(s)$ is integrated TL from shallow traps, S is integrated scintillation, n_{e0} is the initial concentration of electrons captured by activators (Eu^{2+} ions), n_{a} is the concentration of excited activators ($Eu^{3+}*$), $n^{(\mathrm{se})}$ is the concentration of electrons in shallow electron traps, $n^{(\mathrm{sh})}$ is the concentration of holes in shallow hole traps, $N^{(\mathrm{se})}$ is the concentration of shallow electron traps, $N^{(\mathrm{sh})}$ is the concentration of shallow hole traps. Parameters include: f', the effective rate of electron-hole pair production at low temperature, the radiative recombination rate A_{r} of valence-band holes with Eu^{2+} ions; the trapping rates $A^{(\mathrm{se})}$ and $A^{(\mathrm{sh})}$ at shallow electron traps and shallow hole traps, respectively; the non-radiative recombination rate A_{NR} of valence-band holes with electrons in shallow electron traps; and the radiative lifetime τ_{r}. The concentration of holes in deep traps is found to be insensitive to radiation temperature. Optimized parameters are listed in Table 1.

Table 1
Optimized parameters

Parameter	Value
β^{-1}	0.57
α	0.0165
τ_r (s)	0.00116

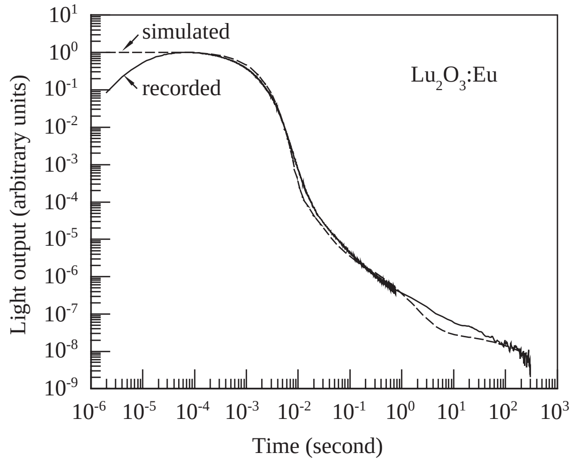

Fig. 2. AG following pulsed X-ray excitation at room temperature. The simulation is based on Eq. (5) and Tables 1 and 2.

4. TL and afterglow (AG)

A trap-depth distribution described by four overlapping Gaussian distributions,

$$G_i \propto \sum_{k=1}^{4} B_k \exp\left[-\frac{(\mathrm{ETRAP}_i - E_k)^2}{2\sigma_k^2}\right] \quad (5)$$

was employed to simulate the TL glow curve produced by an 8 min irradiation at low temperature. The same distribution was adapted to simulate AG, Fig. 2, by adjusting the relative contributions of deep and shallow traps as indicated in Table 2. An additional parameter employed in AG simulations is the branching ratio α between hole-trapping and prompt radiative recombination, listed in Table 1.

A probable explanation for the pronounced reduction in light output and the relative prominence of competing processes at low temperature is a thermal barrier to radiative recombination with estimated activation energy $\Delta E \cong 0.1$ eV,

$$A_r = A_{r0} \exp(-\Delta E/k_B T). \quad (6)$$

Table 2
Optimized parameters for low-temperature TL and room-temperature AG simulations

Parameter	$k=1$ (shallow)	$k=2$ (shallow)	$k=3$ (deep)	$k=4$ (deep)
B_k (TL)	1.00	0.1701	0.1178	0.03876
B_k (AG)	0.1215	0.02067	0.1178	0.03876
E_k (eV)	0.5512	0.7005	0.8973	1.0548
σ_k (eV)	0.03744	0.02484	0.03774	0.06172

5. Discussion

Radiation-enhanced deep hole traps may be associated with anion Frenkel defects, since their Gibbs free energy of formation should be relatively small in the cubic Bixbyite structure of Lu_2O_3 with its abundance of commodious interstitial anion sites [5]. Both holes and electrons are lost to shallow traps at low temperature, with the shallow electron traps also serving as non-radiative recombination centers. Shallow traps may be associated with surface states at grain boundaries.

Acknowledgements

This work was supported in part by NIH Grant Nos. 2R44-DE12785-02A1 and N44-NS-0-2306.

References

[1] R.H. Bartram, D.S. Hamilton, L.A. Kappers, A. Lempicki, J. Lumin. 75 (1997) 183.

[2] R.H. Bartram, D.S. Hamilton, L.A. Kappers, A. Lempicki, J. Glodo, J.S. Schweitzer, C.L. Melcher, Radiat. Effects Defects Solids 150 (1999) 11.

[3] L.A. Kappers, R.H. Bartram, D.S. Hamilton, A. Lempicki, J. Glodo, J. Lumin. 102–103 (2003) 162.

[4] A. Lempicki, C. Brecher, P. Szupryczinski, H. Lingertat, V.V. Nagarkar, S.V. Tipnis, S.R. Miller, Nucl. Instr. and Meth. A 48 (2002) 579.

[5] R. W. G. Wyckoff, Crystal Structures, Vol. 2, 2nd ed, Interscience, NY, 1963–1971, pp. 2–6.