Thermoluminescent response of aluminium oxide thin films subject to gamma irradiation

A. ARRIETA, L. ESCOBAR-ALARCÓN*, E. CAMPS, E. VILLAGRAN, P. R. GONZÁLEZ

Departamento de Física, Instituto Nacional de Investigaciones Nucleares, Apdo. Postal 18-1027, México DF 11801, México

* Corresponding author, e-mail: lea@nuclear.inin.mx

The thermoluminescent (TL) properties of amorphous aluminium oxide thin films (thicknesses as low as 0.3 μm) subjected to gamma (Co-60) irradiation are reported. Aluminium oxide thin films were prepared by laser ablation from an Al_2O_3 target using a Nd:YAG laser with emission at the fundamental line. The films were exposed to gamma radiation (Co-60) in order to study their TL response. Thermoluminescence glow curves exhibited two peaks at 110 and 176 °C. The high temperature peak shows good stability and 30 % fading in the first 5 days after irradiation. A linear relationship between absorbed dose and the thermoluminescent response for doses span from 150 mGy to 100 Gy was observed. These results suggest that aluminium oxide thin films are suitable for detection and monitoring of gamma radiation.

Keywords: Thermoluminescence; Gamma Radiation; Thin Films; Pulsed Laser Deposition

1. Introduction

Materials in thin film form have received great attention mainly because of their singular properties, which may differ significantly from their bulk attributes making them attractive for a wide variety of applications owing to the fact that the film properties can be varied with an appropriate selection of the deposition parameters. In particular, thermoluminescent (TL) properties of thin films have a wide spectrum of potential applications in dosimetry for both ionizing and non ionizing radiation. Among the materials that have been studied for these purposes, the TL response of ZrO_2 films (5 μm thickness) doped with Terbium, produced using the spray pyrolisis technique has been reported [1]. These films were exposed to UV radiation and its TL response has been studied as function of the dose. On the other hand, the TL dosimetric properties of LiF thin films with thickness of 2 μm produced using the electron beam evaporation technique have been also investigated [2]. For thin films with thickness in the sub-micron range, the TL response to UV and beta radiation of aluminium oxide thin films prepared by laser ablation has been reported previously [3, 4]. It is worth noting that TL response in materials in thin film form makes them a very attractive dosimetric tool to address the difficulties associated in the study of dose distributions in interfaces [5, 6].

Pulsed Laser Deposition (PLD) has been extensively used in the last few years to produce complex materials in thin film form owing to its advantages over other deposition techniques. Among these advantages are; the conservation of the stoichiometry on the deposited film, the simplicity of the technique, the possibility to use plastics as substrates and the feasibility to control the thickness of the deposited film [7, 8]. Particularly, PLD has been used successfully to deposit different oxides [9, 10].

The aim of the present work is to report the main TL properties of aluminium oxide thin films prepared by laser ablation with thicknesses as low as 0.3 μm. The TL response was studied as function of the deposition parameters after exposing the thin films to gamma radiation.

2. Experimental Procedure

The experimental set up and the deposition procedure have been reported elsewhere [9]. An Al_2O_3 disk, 99.99% purity, 50 mm diameter and 2 mm thick was used as target. The substrates used in the present experiments were pieces of kapton of 1 cm^2 approximately. Before deposition the kapton substrates were cleaned in distillate water. Target and substrates were placed inside a vacuum chamber with a diffusion pump yielding pressures of the order of $1x10^{-3}$ Pa. The distance target-substrate was fixed at 3.5 cm approximately. The target was rotated during the deposition in order to reduce laser damage and at the same time to increase the irradiated area. The laser used in these experiments was a Q-switched Nd:YAG laser (λ=1064 nm, pulse duration = 28 ns) at 20 Hz repetition rate. The laser beam was focused on the target with a 30 cm focal length spherical lens, its incidence angle was set at 45°. The thin films were grown at room temperature at laser energy densities from 5.0 to 19.0 J cm^{-2}. The deposition time corresponds to approximately 12,000 pulses, resulting in a thin film thickness of approximately 0.3 µm. The film thicknesses were measured with a profilometer.

The TL glow curves were obtained using a Harshaw 4000 TL reader using a cycle composed by a preheat at 50 °C for 5 s followed by an acquisition from 50 to 300 °C at a heating rate of 10 °C s^{-1}. Computerized glow curve deconvolution (CGCD) of a gamma irradiated sample was performed using a heating rate of 2 °C s^{-1} integrating the TL signal from 40 to 300°C. All the TL measurements were performed on a N_2 atmosphere.

3. Results and discussion

Several samples were obtained for the present experiments in order to test the reproducibility of our results. All the films were subjected to the same treatment and characterization. Gamma irradiations were performed using a Co-60 source (E = 1.17 and 1.33 MeV) at absorbed doses from 150 mGy to 100 Gy. All the irradiations and TL measurements were performed at room temperature and the irradiated samples were kept in the dark in order to avoid any influence of the environment light. The readout cycle was repeated after each evaluation in order to guarantee that no remaining TL signal was still present on the sample. With no exception all the samples were completely blank after the first cycle, confirming that no post-evaluation annealing was needed.

The obtained aluminium oxide thin films are amorphous for the deposition conditions used and are slightly deficient in oxygen with a composition that not corresponds exactly to the stoichiometry Al_2O_3. Additionally, the obtained films showed a smooth surface with dispersed splashed particles with diameters ranging from approximately 0.2 to 5 μm [3].

Figure 1 shows the characteristic glow curve of the gamma irradiated films at different absorbed doses from 150 mGy to 100 Gy. In all cases the films were read-out 30 minutes after irradiation. The glow curve exhibited two peaks centred at 110 and 176 °C. As it is observed the TL intensity increases as the gamma dose increases. It is worth mentioning that the replicate TL results from every sample irradiated with gamma radiation were always better than 5 %, nevertheless differences in sensitivity up to a factor of 2 were observed within five samples studied under the same conditions. In order to verify that the TL signal observed on the irradiated samples was not arising from the kapton substrates, pieces of kapton without film were irradiated in the same conditions and no TL response was observed.

The dependence of the TL response as a function of the energy density used for thin film deposition is shown in figure 2. For energy densities lower than 12 J cm^{-2} the TL intensity starts increasing gradually and then the TL intensity increases almost 1 order of magnitude for energy densities from 12 J cm^{-2} to 19 J cm^{-2}. This behaviour could be explained in terms of the fact that a more energetic and dense plasma plume is obtained when the energy density delivered to the target is increased; therefore a more intense bombardment by energetic species during deposition favours the TL response as it can produce a higher number of defects. Is worth mentioning that the as grown samples show thermoluminescence response read after the deposition process. This signal can be attributed to defects that are induced during the deposition process as well as due to the irradiation of the growing film by x-rays, UV, ions and electrons presents in the ablation plasma.

The typical TL response, considered as the integrated signal from the high temperature peak of the gamma irradiated films, as a function of the absorbed dose is shown in figure 3. This reveal a linear relationship between dose and TL response in the dose range studied and suggest that the high temperature peak could be used as the dosimetric peak.

In order to study the fading of the TL response, a set of 5 samples were irradiated at an absorbed dose of 40 Gy and then were read out after 1 to 5 days. In general terms it was observed that the 176 ºC peak shows good stability whereas the low temperature peak fades completely in approximately 10 hours. Figure 4 show that the TL signal fades 3.2 % in the first day, fading to approximately 30% after 5days.

The TL parameters obtained by gamma irradiation of the amorphous aluminium oxide thin films using the CGCD method [11] revealed a second order kinetics (b = 2.0) and an activation energy of 1.2 eV for the 176 ºC peak while the low temperature peak exhibited an activation energy of 0.7 eV.

4. Conclusions

The results presented in this work show that is possible to obtain materials in thin film form with thicknesses as low as 0.3 μm which exhibit TL response to gamma radiation. The stability of the 176 °C peak and the linear relationship between dose and TL response for gamma radiation suggest that aluminium oxide thin films are potentially attractive in radiation dosimetry as ultra-thin detectors for this kind of radiation.

Acknowledgements

This work has been partially supported by CONACYT México, under grant 46344.

References

[1] J. Azorin, T. Rivera, C. Falcony, E. Martinez, M. García; Rad. Prot. Dosim., **85**, 317 (1999).

[2] C.L.P. Mauricio, R.A. Nunes, M.H.P. Mauricio, L.C. Scavarda do Carmo; Rad. Prot. Dosim., **84**, 223 (1999).

[3] L. Escobar-Alarcón, E. Villagrán, E. Camps, S. Romero, J. E. Villarreal-Barajas and P.R. González; Thin Solid Films, **433,** 126-130 (2003)

[4] J. E. Villarreal-Barajas, L. Escobar-Alarcón, P. R. González, E. Camps, and M. Barboza-Flores; Radiation Measurements, **35**, 355-359 (2002)

[5] P. Bilski, B. Olko, B. Burgkhardt, E. Piesc, Radiat. Meas., **24**, 445 (1995).

[6] J.E. Villarreal-Barajas, J.P. Darley, M.W. Charles, Rad. Prot. Dosim., **85**, 501 (1999).

[7] H. Sankur, R. Hall, Appl. Opt., **24**, 3343 (1985).

[8] L. Ponce, M. Fernández-Guasti, E. Jiménez, E. Haro-Poniatowski, Rev. Méx. Fis., **40**, 798 (1994).

[9] L. Escobar-Alarcón, E. Haro-Poniatowski, M.A. Camacho-Lopez, M. Fernández-Guasti, J. Jimenez-Jarquin y A. Sanchez-Pineda, Surface Engineering, **15**, 411 (1999).

[10] L. Escobar-Alarcón, E. Haro-Poniatowski, M. Fernández-Guasti, C. N. Afonso, A. Perea, Appl. Phys. A, **69,** 949 (1999).

[11] R. Chen, Kinetics of thermoluminescence glow peaks. in: *Thermoluminescence and thermoluminescent dosimetry*, Y.S. Horowitz (Ed), CRC Press, Boca Raton (1984).

Figure Captions

Figure 1. TL glow curves of gamma irradiated aluminium oxide thin films at different doses.

Figure 2. TL intensity as a function of the laser energy density used for thin film deposition.

Figure 3. TL response of the irradiated films as function of the gamma absorbed dose. The solid line is guide to the eye.

Figure 4. TL signal fading as a function of the time.

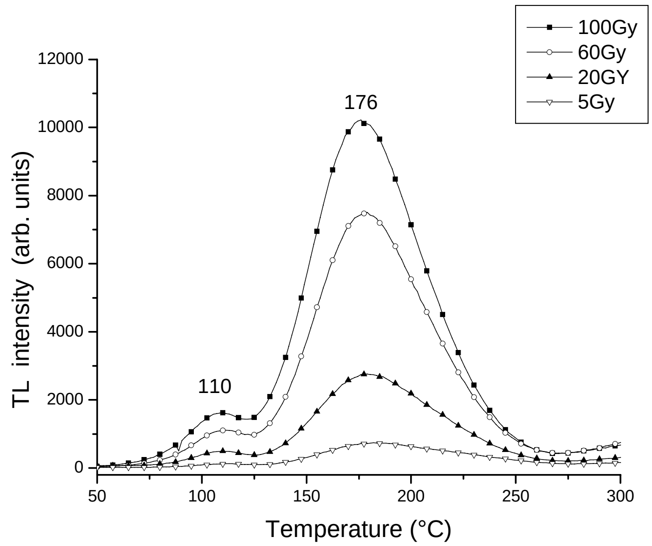
100Gy
60Gy
20GY
5Gy
176
110
TL intensity (arb. units)
Temperature (°C)
0
2000
4000
6000
8000
10000
12000
50
100
150
200
250
300

Figure 1

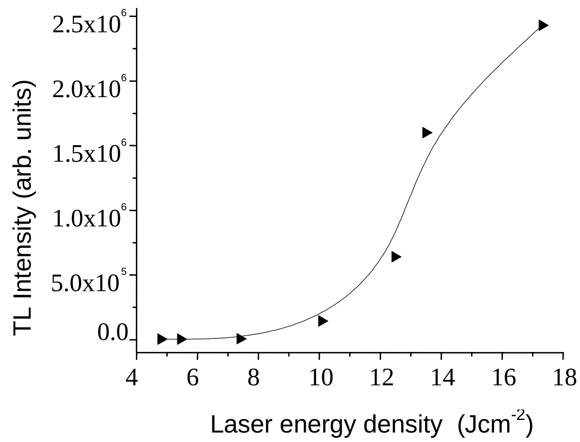
TL Intensity (arb. units)
2.5x10^6
2.0x10^6
1.5x10^6
1.0x10^6
5.0x10^5
0.0
4
6
8
10
12
14
16
18
Laser energy density (Jcm^-2)

Figure 2

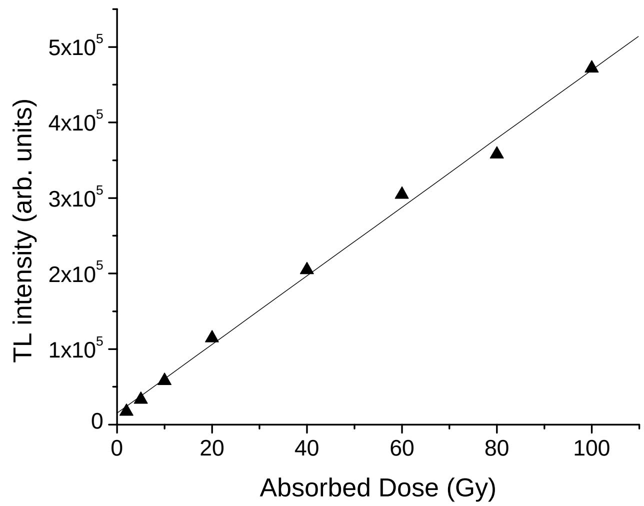
TL intensity (arb. units)
0
1x10^5
2x10^5
3x10^5
4x10^5
5x10^5
0
20
40
60
80
100
Absorbed Dose (Gy)

Figure 3

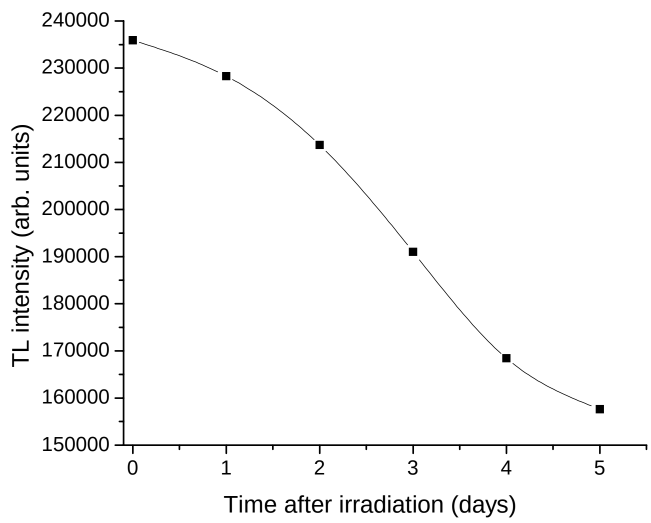
240000
230000
220000
210000
200000
190000
180000
170000
160000
150000
TL intensity (arb. units)
0
1
2
3
4
5
Time after irradiation (days)

Figure 4