Thermoluminescence response of aluminum oxide thin films to beta-particle and UV radiation

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Aluminium oxide thin films (0.3 μm) were grown by laser ablation from an Al_2O_3 crystal disk. The thin films were deposited on pieces of silicon (100) wafers. The structural characterization revealed the growth of an amorphous material. Surface morphology of the obtained thin films show a smooth surface with dispersed splashed particles with diameters ranging from approximately 0.2 μm to 5 μm. The films were exposed to UV (254 nm) and beta-particle radiation (^{90}Sr-^{90}Y). Thermoluminescence glow curves exhibited two peaks centered at 95 and 162 °C for UV irradiation. For beta-particle irradiation the thermoluminescence glow curve shows only the presence of the high temperature peak. The 162 °C peak shows good stability and 10% fading in the first 4 days after irradiation. A linear relationship between absorbed dose and the thermoluminescence response up to 20 Gy was observed for beta-particle irradiation. The thermoluminescence parameters obtained showed a second order kinetics and an activation energy of 1.2 eV for the 162 °C peak. These properties make aluminum oxide thin films potentially attractive as an ultra-thin dosimeter for UV and beta-particle radiation.

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1. Introduction

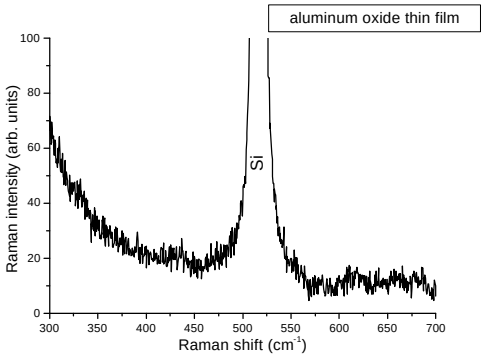
Materials in thin film form have received great attention mainly because of their singular properties, which may differ significantly from their bulk attributes making them attractive for a wide variety of applications. Additionally, thin film properties can be varied with an appropriate selection of the deposition parameters as well as using suitable post deposition treatments. In this way it is possible to grow materials with specific properties for tailored applications. In particular, thermoluminescence (TL) properties of thin films have a wide spectrum of potential applications in dosimetry for both ionizing and non ionizing radiation. The interest to study the TL response of thin films has been motivated by their importance in the measurements of absorbed doses produced by weakly penetrating radiation as well as, in the study of dose distributions in interfaces [1-4]. 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 [5]. 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 [6]. Additionally, the TL response of sodium chloride thin films prepared by laser ablation has been reported previously [7]. However, work on the TL response of films with thickness in the sub-micron range has not been reported. It is worth noting that TL response in these films make them a very attractive dosimetric tool to address the difficulties associated with the measurements of depth dose distributions produced by weakly penetrating radiation over very thin layers such as that of the basal cells in the skin [8].

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 [9-11]. Particularly, PLD has been used successfully to deposit complex oxides [12,13].

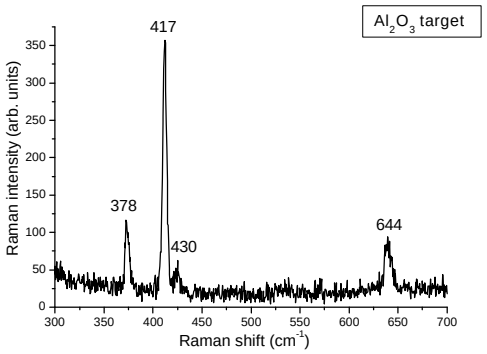
The aim of the present work is to report the main TL properties of aluminum oxide thin films prepared by PLD with thicknesses as low as 0.3 μm.

2. Experimental Procedure

The experimental set up and the deposition procedure have been reported elsewhere [14]. An Al_2O_3 crystal disk 1 mm thick and 5 mm diameter obtained from the Urals Polythecnical Institute, Ekaterinburg, Russia (TLD 500 K) was used as target. The films were deposited on pieces of silicon (100) wafers previously cleaned in an acetone and ethylic alcohol using an ultrasonic bath. Target and substrates were placed inside a vacuum chamber with a diffusion pump yielding pressures of $8x10^{-6}$ Torr. The distance between them was fixed at 35 mm approximately. The target was rotated during the deposition in order to



a)



b)

Figure 1. (a) Raman spectrum of the aluminum oxide thin film prepared by PLD, (b) Raman spectrum of the Al_2O_3 target used for the PLD experiments.

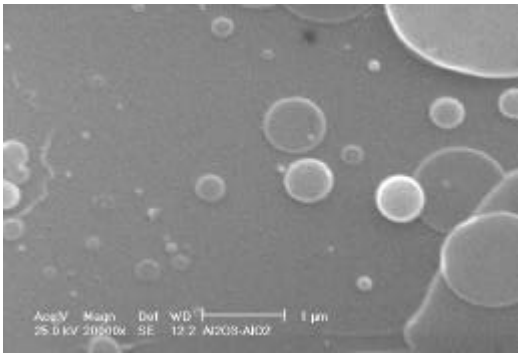


Figure 2. Micrograph of an aluminum oxide thin film prepared by pulsed laser deposition.

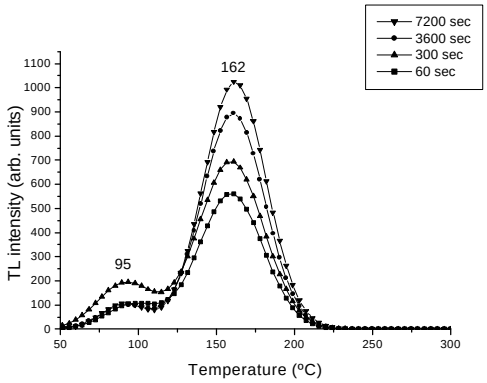


Figure 3. TL glow curves of UV irradiated aluminum oxide thin films at different times.

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 200 mm focal length spherical lens, its incidence angle was set at 45°. The thin films were grown at room temperature at a mean power densities close to 10^8 W/cm^2. The deposition time corresponds to approximately 12,000 pulses, resulting in a thin film
thickness of approximately 300 nm. The film thicknesses were determined with a profilometer (Sloan Dektak II).

The structural properties of the synthesized films were studied by Raman spectroscopy and X-ray diffraction (XRD). Raman spectroscopy measurements were performed at room temperature in air with a Spex 1403 double monochromator using the 514.5 nm line of an Argon-ion laser (Laser Ionics) as excitation source at a power level of 100 mW in a backscattering configuration. The signal was detected with a photomultiplier and a standard photon counting system. The X-ray diffraction measurements were performed in a Siemens D-5000 diffractometer with a CuK_α radiation source (λ_k = 1.5406 Å).

The surface morphology of the films was analyzed using a Phillips XL30 scanning electron microscope. Surface chemical analysis was performed using energy dispersive spectrometry (EDS).

The TL glow curves were obtained using a Harshaw 4000 TL reader. The glow curves were obtained 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 an UV irradiated sample was performed using a heating rate of 2 °Cs^{-1} integrating the TL signal from 40 to 300°C. All the TL measurements were performed on a N_2 atmosphere. 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.

3. Results and discussion

Two series of experiments were performed, one to investigate the thermoluminescence response of the aluminium oxide thin films irradiated with UV radiation and the other to investigate the TL response due to beta-particle radiation. For the UV irradiation a low pressure Hg UV lamp with a main emission line at 254 nm was used. Beta-particle irradiations were performed using a ^{90}Sr-^{90}Y source ($E_{max.}$= 2.28 MeV). 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 films were irradiated using the same experimental set up.

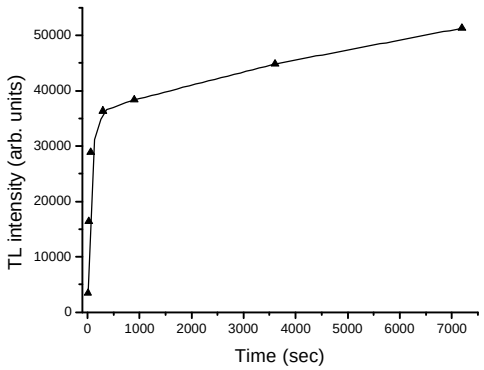


Figure 4. TL response of the UV irradiated films as function of the irradiation time. The solid line is guide to the eye.

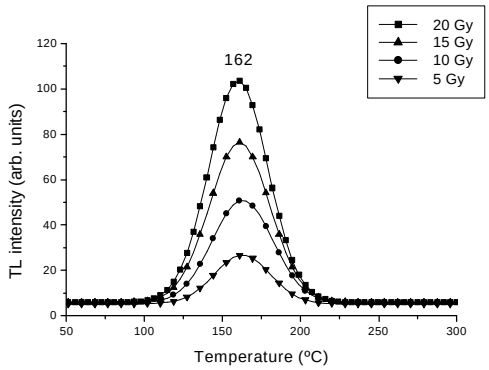


Figure 5. TL glow curves of beta-particle irradiated thin films at doses from 5 to 20 Gy.

In figure 1(a) the Raman spectrum corresponding to an aluminum oxide thin film is presented, the absence of peaks indicates that the material is amorphous. This can be due to the low mobility of the particles impinging the cold substrate. The corresponding x-ray diffraction pattern showed no diffraction peaks consistent with the Raman results. As a reference the Raman spectrum presented in figure 1(b) corresponds to the target which clearly shows a crystalline structure. Therefore XRD and Raman spectroscopy measurements revealed the growth of an amorphous material.

Surface morphology of the obtained thin films was analyzed using a scanning electron microscope. The films show a smooth surface with dispersed splashed particles with diameters ranging from approximately 0.2 μm to 5 μm as shown in figure 2. These particles have been ejected from the target due to surface heating above the melting point with the subsequent fusion of the surface. Furthermore the composition of the splashed particles measured using EDS showed that the splashed particles have very similar composition to the underlying film. The ratio O/Al was approximately 1.3 that corresponds to an aluminum oxide.

Figure 3 shows the characteristic glow curve of the UV irradiated films at different times. The UV irradiation time of the samples span from 60 seconds to 2 hours (read-out 30 minutes after irradiation). The curve exhibited two peaks centered at 95 and 162 °C. Similar results were obtained when Al_2O_3:C crystals were irradiated with UV radiation showing peaks at 58 and 184 °C [15]. The replicate TL results from every sample irradiated with UV was always better than 5 %, nevertheless differences in sensitivity up to a factor of 2 were observed within five samples studied. In order to verify that the TL signal observed on the irradiated samples was not arising from the Si substrate, pieces cut from the same Si wafer were irradiated in the same conditions and no TL response was observed

The typical TL response (considered as the integrated signal from the high temperature peak) of the UV irradiated films as function of the irradiation time is shown in Figure 4. From this figure it is evident that this material exhibits a non-linear growth of TL intensity with dose (irradiation time) approaching saturation at high doses. This is a sub-linear like dose dependence effect. The TL signal starts increasing at a high rate for low doses and then increase gradually much more slower until it reaches a stable value. This behavior may suggests the existence of a TL mechanism that involves at least two trap levels, one acting as a competitor and the other as the active trap [16]. The fading of the 162 ºC peak was of almost 30 % in the first 6 hours, reducing to approximately 50% after 4 days. On the other hand the low temperature peak fades completely in 10 hours.

In figure 5 the glow curves obtained for beta-particle irradiation at doses ranging from 2.5 to 20 Gy are showed. As can be observed these glow curves show only the presence of the high temperature peak (162 ºC) observed when the sample was irradiated with UV (see figure 3). It is worth mentioning that the absence of the low temperature peak (95 ºC) on these beta-particle irradiated samples may indicate that the low temperature peak observed on the UV irradiated samples is produced essentially by a photo-transfer process.

The 162 ºC peak shows good stability and 10% fading in the first 4 days. In this case the material shows a linear relationship between dose and TL response in the dose range studied, this is displayed in Figure 6. It is worth mentioning that for doses below 2.5 Gy the TL response was very poor. As for the UV irradiations the TL response reproducibility was always better than 5 % and differences in sensitivity up to a factor of 4.

The TL parameters obtained by UV irradiation of the amorphous aluminum oxide thin films using the CGCD method [17,18] revealed a second order kinetics (b = 2.0) and an activation energy of 1.2 eV for the 162 ºC peak while the low temperature peak exhibited an activation energy of 0.7 eV.

Additionally, thermoluminescence measurements of not-irradiated samples (carried out after laser ablation deposition) revealed that the films have an intrinsic TL

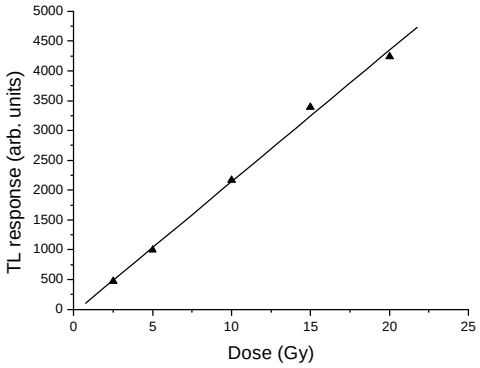


Figure 6. TL response of the beta-particle irradiated films as function of the dose.

response, this signal can be attributed to defects that are induced during the deposition process.

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 UV and beta-particle radiation. These results suggest that the 162 °C peak can be used as a dosimetric peak. The linear relationship between dose and TL response for beta-particle radiation make aluminum oxide thin films suitable as ultra-thin dosimeter for beta-particle radiation.
A more detailed characterisation of the thin films as well as the effects on the thin film properties own to the deposition conditions is under investigation. This investigation may lead to an explanation of the differences observed in the sensitivity of the aluminum oxide thin films.

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References

[1] P. Bilski, B. Olko, B. Burgkhardt, E. Piesc, Radiat. Meas., **24**, 445 (1995).
[2] J. Bhom; et al. 1998. Dosimetry of weakly penetrating radiation, PTB-Dos-30, Braunschweig.
[3] J.E. Villarreal-Barajas, J.P. Darley, M.W. Charles, Rad. Prot. Dosim., **85**, 501 (1999).
[4] J.E. Villarreal-Barajas, J.P. Darley, M.W. Charles, Rad. Prot. Dosim., **85**, 507 (1999).
[5] J. Azorin, T. Rivera, C. Falcony, E. Martinez, M. García, Rad. Prot. Dosim., **85**, 317 (1999).
[6] C.L.P. Mauricio, R.A. Nunes, M.H.P. Mauricio, L.C. Scavarda do Carmo, Rad. Prot. Dosim., **84**, 223 (1999).
[7] A. Arrieta, S. Mera, R. Diamant, M. Fernández-Guasti, R. Sosa, L. Escobar-Alarcón, A.F. Muñoz, E. Haro-Poniatowski, Appl. Phys. A, **69**, s491 (1999).
[8] (ICRU Report 56, 1996).
[9] H. Sankur, R. Hall, Appl. Opt., **24**, 3343 (1985).
[10] J. T. Cheung, History and fundamentals of Pulsed Laser Deposition, in: D.B. Chrisey, G.K. Hubler (Eds.) *Pulsed Laser Deposition of Thin Films*. (Wiley, New York, 1994).
[11] L. Ponce, M. Fernández-Guasti, E. Jiménez, E. Haro-Poniatowski, Rev. Méx. Fis., **40**, 798 (1994).
[12] L. Escobar-Alarcón, E. Haro-Poniatowski, M. Fernández-Guasti, C. N. Afonso, A. Perea, Appl. Phys. A, **69,** 949 (1999).
[13] C. Julien, E. Haro-Poniatowski, M.A. Camacho-López, L. Escobar-Alarcón, J. Jiménez-Jarquín, Materials Science and Engineering B, **72,** 36 (2000).
[14] L. Escobar-Alarcón, E. Haro-Poniatowski, M.A. Camacho-López, M. Fernández-Guasti, J. Jiménez-Jarquín y A. Sánchez-Pineda, Surface Engineering, **15**, 411 (1999).
[15] R. Pérez-Salas, R. Aceves, R. Meléndrez, M. Barboza-Flores, L. Pashchenko, Appl. Phys. Lett., **63**, 894 (1993).
[16] R. Chen, S.W.S. Mckeever, *Theory of Thermoluminescence and related phenomena*, (World Scientific, Singapore, 1997).
[17] R. Chen, Kinetics of thermoluminescence glow peaks. in: *Thermoluminescence and thermoluminescent dosimetry*, Y.S. Horowitz (Ed), CRC Press, Boca Raton (1984).
[18] P.R. González, J. Azorin, Radiat. Phys. and Chem., *in press*.