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To cite this article: J Garcés *et al* 2017 *J. Phys.: Conf. Ser.* **792** 012007

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Thermoluminescent response of C-modified Al₂O₃ thin films deposited by parallel laser ablation plasmas

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Abstract. Aluminium oxide thin films modified with different amounts of carbon were prepared using a parallel laser ablation plasmas configuration. The effect of the amount of carbon incorporated in the films on their compositional, morphological, structural, and thermoluminescent properties was studied. The results showed that films with different C content, from 11 to 33 at. %, were obtained. The structural characterization revealed the growth of an amorphous material. Surface morphology of the obtained thin films showed smooth surfaces. The films were exposed to UV and gamma radiation (Co-60) in order to study their thermoluminescence response. The results tend to indicate that carbon incorporation into the alumina favours the increase of a high temperature TL peak.

1. Introduction

Thermoluminescence (TL) properties of thin films have a wide spectrum of potential applications in dosimetry for both ionizing and nonionizing radiation. The interest to study the TL response of thin films has been motivated because of their importance in the measurements of absorbed doses from weakly penetrating radiation as well as in the study of dose distributions in interfaces [1]. Particularly, Al₂O₃ is an important TL material studied for its application as a radiation dosimeter due to its high thermal and chemical stability and low effective atomic number [2]. It has been reported elsewhere [3] that the improvement of the TL response of Al₂O₃ can be achieved by the intentional introduction of defects such as oxygen vacancies and impurity centers formed by carbon. In particular, doping of α -Al₂O₃ with carbon improves its TL response resulting in a material with good dosimetric properties with potential applications in various areas of dosimetry, such as personnel, neutron, space, medical, environmental, and emergency dosimetry [4]. Although C doped Al₂O₃ is being used and investigated extensively, the role of carbon in rendering this phosphor remarkably sensitive towards radiation is not well understood [5]. Therefore, a lot of work is still necessary to investigate this issue.



Among the thin film deposition techniques, laser ablation has been employed to prepare thin films of carbon nitride [6], as well as aluminum oxide with thermoluminescent response when subjected to UV, gamma and beta radiation [7, 8, 1]. In particular, the versatility of the laser ablation technique allows the possibility of implementing different alternative configurations in order to improve some characteristics of the deposits as has been reported elsewhere [9, 10]. In the present work, the preparation of aluminium oxide thin films modified with different amounts of carbon, with TL properties, using a parallel laser ablation plasmas deposition configuration is reported.

2. Experimental Procedure

Thin films were deposited by the combination of two laser ablation plasmas produced sequentially. The plasma plumes were generated by laser ablation using the third harmonic of a Nd:YAG laser with emission at 355 nm and 10 ns pulse duration at a frequency of 10 Hz. The laser beam was focused onto the targets with a 50-cm focal length spherical lens. The targets were disks of high purity (99.99%) of aluminum oxide (Al_2O_3) and carbon (50 mm diameter and 2 mm thick). The laser fluence delivered on both targets was kept constant at 8.2 J/cm^2 . By varying the ratio of the ablated areas of carbon and alumina (AA_C/AA_A), thin films modified with different amounts of carbon were deposited. The target to substrate distance was set at 5 cm and the deposition time was 60 min. Thin films were deposited at room temperature onto glass and silicon substrates (1x1 inch). Determination of the elements presents in the deposits as well as their bonding features were done by X-ray Photoelectron Spectroscopy (XPS) using a Jeol JPS 9200 XPS. The structural properties of the synthesized films were studied by Raman spectroscopy using an HR LabRam 800 system equipped with an Olympus BX40 confocal microscope and a Nd:YAG laser (532 nm). The surface morphology of the films was analyzed using a Phillips XL30 scanning electron microscope. The TL glow curves were obtained using a Harshaw 4000 TL reader using a cycle composed by a preheat at $50 \text{ }^\circ\text{C}$ for 5 s followed by an acquisition from 50 to $300 \text{ }^\circ\text{C}$ at a heating rate of $10 \text{ }^\circ\text{C s}^{-1}$. The TL measurements were performed under 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. For the UV irradiation a low pressure Hg UV lamp with main emission at 254 nm and an energy density of approximately $1350 \text{ } \mu\text{W/cm}^2$ was used. Gamma irradiations were performed using a Co-60 source ($E = 1.17$ and 1.33 MeV) at an absorbed dose of 10 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 environmental light.

3. Results and Discussion

3.1 Compositional characterization

The XPS results showed that the elements present in the film were aluminium, oxygen and carbon. Figure 1a shows the carbon content as a function of the ratio of the carbon to alumina ablated areas (AA_C/AA_A). It is observed that the carbon content in the film follows a monotonic behaviour, increasing its content as the AA_C/AA_A increases. In this way it was possible to vary the carbon content from 11 to 33 at.%.

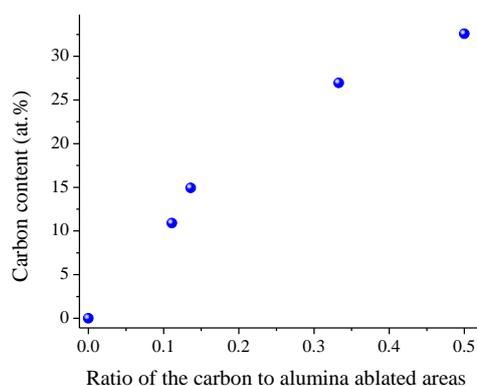


Figure 1. Carbon content as a function of the ratio of the carbon to alumina ablated areas

Figure 2 shows the high-resolution XPS spectrum of the O1s region of the samples prepared with different carbon content. The peak at 531.5 eV observed in the spectrum of the sample without carbon is assigned to O1s in Al₂O₃. As the carbon is incorporated into the film the position of this peak is shifted to 532 eV revealing that oxygen is bonded to carbon atoms forming O=C bonds.

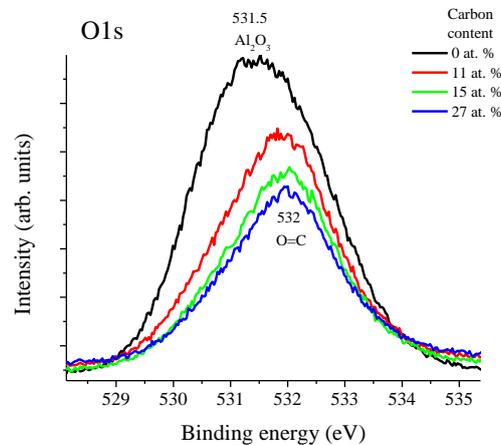


Figure 2. High-resolution XPS spectrum of the O1s region

3.2 Raman characterization

In figure 3 the Raman spectrum corresponding to an aluminium oxide thin film with a carbon content of 33.0 at.% is presented. The peaks at 300, 521 and 900 cm⁻¹ correspond to the silicon substrate. The broad band from 1000 to 1800 cm⁻¹ is due to carbon. The absence of peaks characteristics of alumina indicates that the material is amorphous.

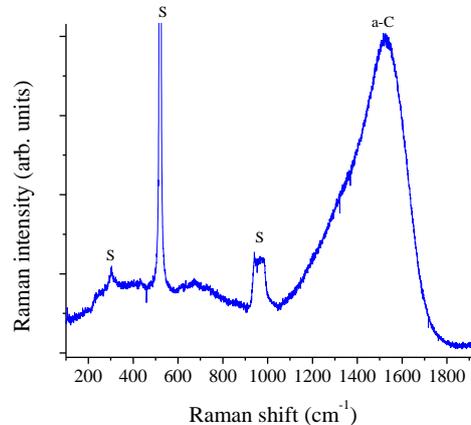


Figure 3. Raman spectrum of the sample with a carbon content of 33 at.%

Figure 4 shows the region from 1000 to 1800 cm⁻¹ which is very similar to those previously reported for DLC films being characterized by an asymmetric band composed of two sub-bands in different proportions [11]. One of them in the range of 1560-1600 cm⁻¹, the so-called G band associated with the in-plane stretching motion of pairs or chains of C sp² bonded atoms. The second band around 1350 cm⁻¹, the D band, is associated with a breathing mode of six fold aromatic rings and only becomes active in presence of disorder [11]. In order to perform the data analysis, the Raman spectra were fitted using a Breit-Wigner-Fano (BWF) line shape for the G peak and a Lorentzian line shape for the D peak. The interpretation of the Raman results was performed following the model proposed by Ferrari [12], which give information about the content of sp³ bonded carbon (diamond-like degree). The obtained results were: G position = 1560 cm⁻¹ and I_D/I_G = 1.1 which according to the Ferrari's model correspond to a C-sp³ content of approximately 30 %.

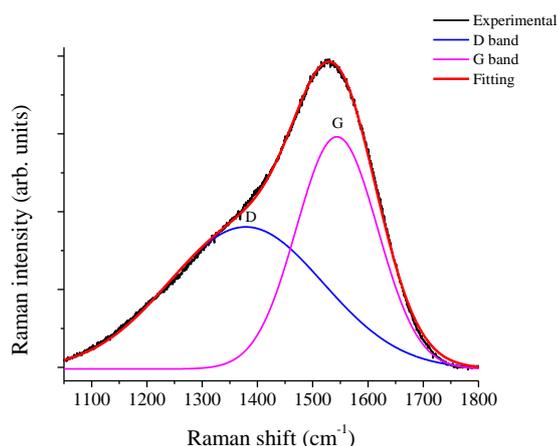


Figure 4. The 1000 to 1800 cm^{-1} Raman spectrum region showing the fitting performed

3.3 Surface morphology

The SEM images of films without carbon and with the maximum content of carbon are shown in figures 5a and 5b respectively. It is clearly seen that films with very smooth surfaces are obtained suggesting no effects on the surface morphology due to the carbon incorporation.

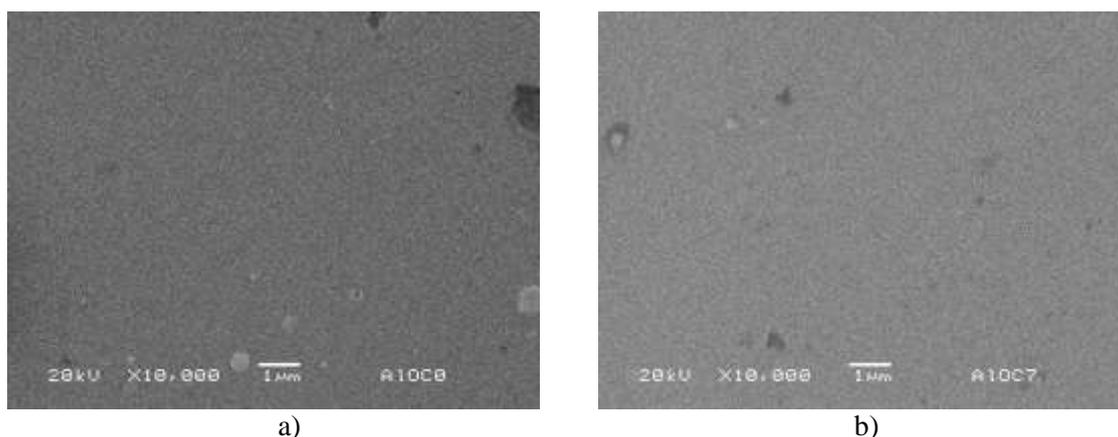


Figure 5. SEM images of films, a) without carbon and b) with a carbon content of 33 at.%

3.4 TL response

Two series of experiments were performed, one to investigate the thermoluminescence response of the C-modified aluminium oxide thin films subjected to UV radiation and the other to investigate the TL response due to gamma radiation. 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 environmental light. It is worth noting that TL measurements of non-irradiated samples revealed that the films have an intrinsic TL response attributed to defects induced during the deposition process.

Figure 6a shows the characteristic TL glow curves of UV irradiated films with different carbon content. The curves exhibit very broad peaks centered approximately at 156 °C for the sample without carbon and 183 °C for the C-modified alumina films. The differences observed in the peak positions can be attributed to the presence of carbon and the shift is indicative of the presence of impurities or to a slightly different composition of the materials used in each case. The typical TL response (considered as the integrated signal of the TL band) of the UV irradiated films as a function of the irradiation time is shown in Figure 6b. From this figure two main points can be emphasised: a) clearly, the film without carbon exhibits the higher TL response and for carbon contents greater than 15 at.% the TL response remains almost constant; b) it is evident that the prepared material present different behaviours of the TL intensity with dose (irradiation time) depending on the carbon content, the

sample without carbon and with 11 at.% show and increase for irradiation times up to 30 min approaching saturation at higher doses. This is a sublinear-like dose dependence effect in which the TL signal starts increasing at a high rate for low doses and then increase gradually slower until it reaches a final value. The films with carbon contents higher than 15 at.% show a monotonic behavior increasing the TL signal as the irradiation time increases.

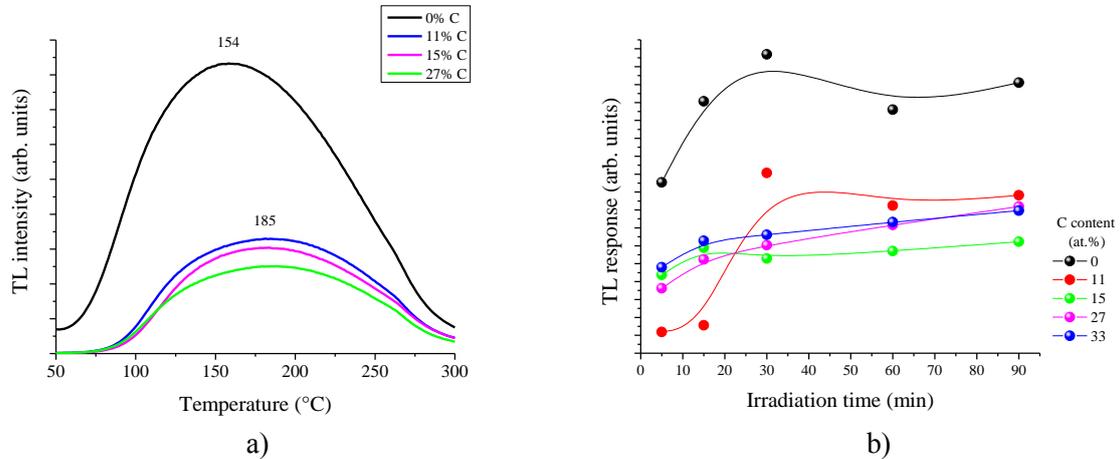


Figure 6. a) Typical TL glow curves of UV irradiated films, b) TL response as function of the UV irradiation time

In Figure 7a the glow curves of the films irradiated at a Co-60 dose of 10 Gy are shown. Two clear bands peaking at 170 and 280 °C are observed. These glow curves have very different shapes compared with the curves obtained under UV irradiation, suggesting that different kind of traps are activated depending on the kind of radiation used. It is worth mentioning that deep traps are more useful for dosimetry purposes due to their higher stability.

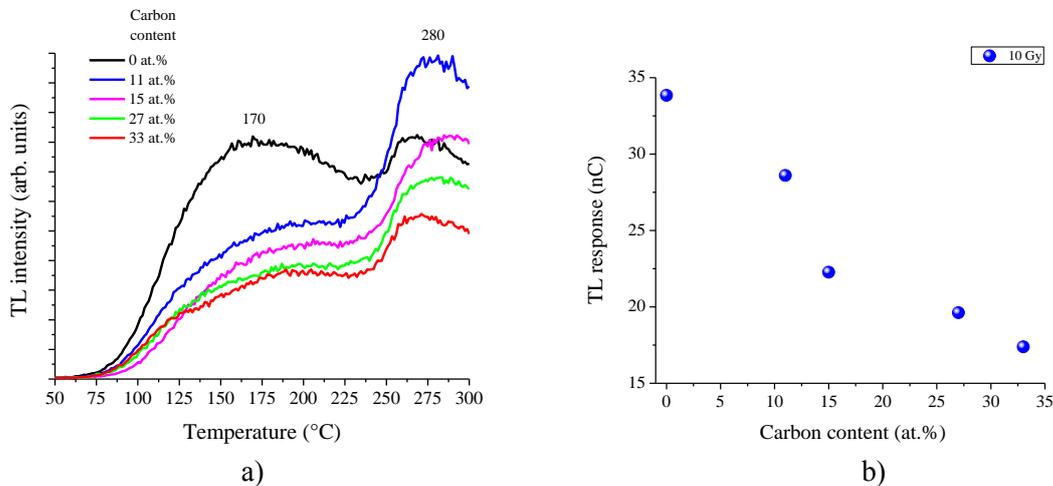


Figure 7. a) glow curves of the films irradiated with Co-60 gammas, b) TL response as a function of the carbon content of samples irradiated at 10 Gy of gamma radiation

Additionally, it is clearly seen that carbon content favors the increase of the high temperature TL peak. In this case the material shows a linear relationship between carbon content and TL response for the dose studied, as it is observed in Figure 7b. It is worth noting that for samples containing carbon the TL response decreases as the C content increases.

4. Conclusions

The results presented in this work show that it is possible to obtain C-modified Al_2O_3 thin films which exhibit TL response to UV and gamma radiation. The deposition configuration employed in which the combination of two laser ablation sequentially produced plasmas allows to control the incorporation of carbon in a simple setup. In spite of the fact that unmodified alumina films exhibit a higher TL

response, the obtained results seem to indicate that carbon incorporation into the alumina favours the increase of a TL peak of high temperature indicative of the presence of deep traps more useful for dosimetric purposes due to their better stability. A more detailed characterization of the thin films as well as the effects on the thin film properties depending on the deposition conditions in order to optimize the TL response is under investigation.

5. Acknowledgements

This research project was partially supported by CONACYT under the contract CB-240998

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