The Characterizations of La$_2$Ti$_2$O$_7$ Thin Films Deposited by Pulsed Laser Deposition at Different Annealing Temperatures

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Abstract: Lanthanum titanium oxide thin films are sometimes used in high-temperature environments. Therefore, it is worth paying attention to the thermal stability of the La$_2$Ti$_2$O$_7$ films. La$_2$Ti$_2$O$_7$ target was made through traditional solid-state reaction way to study the effect of substrate temperature on the characteristics of LTO thin films; A Set of lanthanum titanium oxide thin films has been deposited on to Si (100) substrate through Pulsed laser deposition at different annealing temperatures. The results of X-ray diffraction indicated that the prepared LTO thin films at temperatures up to 700°C are amorphous, while the profilometer Dektak-XT conducted to determine the thickness and roughness of La$_2$Ti$_2$O$_7$ films. The obtained result pointed that the thin film thickness decreased by increasing annealing temperature linearly, and the roughness was inversely proportioning to the increasing of substrate temperature. The value of the lowest roughness equal to 12.28 nm for the thinner film with a thickness of 253.46 nm, while the highest roughness was found to be 14.74 nm for the thicker film at 323.05 nm, which were deposited at 700°C and 500°C respectively, therefore it has been remarked that the annealing temperature influenced the morphology, thickness, and roughness of the LTO thin film.

Keywords: La$_2$Ti$_2$O$_7$, Thin Films, PLD, Perovskites, Annealing Temperature

1. Introduction

In the past decades, many experimental and theoretical studies were blessed to the study of perovskite materials because of their interesting ferroelectric, Piezoelectric, and dielectric properties [1-3]. These interesting properties make LTO a material of choice for extensive technological applications such as amplifiers and aerospace applications [4, 5], Transducers device [6], sensors, wireless communications [7-9], And Built Infrastructure applications [10]. La$_2$Ti$_2$O$_7$ is a part of the layered perovskite family [11], which has the highest Curie temperature with excellent piezoelectric and electro-optic properties. This makes LTO thin films a powerful candidate for a variety of applications in electrical and optical devices [12, 13]. Therefore, LTO thin films have been grown by several techniques including spray pyrolysis method deposition [14], RF magnetron sputtering [15], electron beam evaporation [16], a polymeric organic solution and pulsed laser deposition (PLD) [17, 18]. The choice of the technique and the preparation conditions are the critical point of a research topic since the structural, optical and electrical properties of these films are strongly associated with the elaboration process and operating conditions [19]. Indeed, PLD considered as one of the most promising techniques for thin films synthesis, due to its unique advantages as high
reproducibility, control of the films growth rate, low impurity concentration in the composition of deposited films and possibility to use different substrate materials [20, 21]. In this article, we evaluated the effect of the annealing temperature on the crystalline status and morphology of the LTO films deposited by Pulsed Laser Deposition on Si substrate under different temperatures.

2. The Experimental Procedure

Lanthanum titanium oxide powder was prepared using traditional solid-state method, namely by mixing a stoichiometric amount 2:1 of (Aldrich 99.99%) TiO$_2$ and La$_2$O$_3$ (Aldrich 99.99%) powder. After grindings using a mortar and pestle, the mixture was successively calcined at 1400°C for 4h using High-Temperature Furnace (Delta Power Controls). In order to improve the synthesis efficiency, the calcined product was pelleted using uni-axial pressing (Kimaya Engineers). Then the formed pellet heat-treated in air at 1100 °C for 8 hours with immediately grinding. For the deposition process, a 45.8% density target was obtained by uni-axial pressing of the LTO powder under 20 MPa followed by a sintering at (1350 °C) for 10 hours in air. Then the sintered pellet characterized by a (Rigaku) X-ray diffractometer and ultimately the LTO pellet placed as a target in the film deposition process. Before the deposition, Si substrates were cleaned with RCA to eliminate all irrelevant objects from the surface and finally the substrate diced into (4 cm$^2$) pieces. LTO thin film deposited on (100) Si substrates using a KrF excimer laser (COHERENT LMC; $\lambda$=248 nm; t ~ 20 ns) with constant laser frequency (3 Hz) and fluency (200 mJ), together with a target substrate distance of 5 cm for 85 minutes under vacuum pressure $\sim$10$^{-6}$ mbar at different annealing Temperatures 500, 600 and 700°C, Separately. The structural analysis taken using the Rigaku X-ray diffractometer technique (Cu Kα radiation), while the actual thickness and the roughness of the thin films were obtained using a Bruker profilometer Dektak- XT.

3. Results and Discussions

3.1. Sample Characterization

The XRD pattern of the LTO target at room temperature was analyzed and determined using XPert High Score Plus software which was classified into a monoclinic structure with the lattice a: 13.0150 (Å), b=5.5456 (Å) and c=7.8170 (Å) which are fairly consistent with those stated in the literature as exhibited in figure 2 [19]. Figure 3 presents the x-ray diffraction patterns for La$_2$Ti$_2$O$_7$ films annealed at 500, 600 and 700°C on a Si substrate. The x-ray diffraction pattern exhibits an amorphous peak and no fine spectral peak distribution for the LTO thin films, which means that the film is amorphous with two peaks of the Si substrate [22].

3.2. Thickness and Roughness Measurement:

The thickness of the LTO thin films deposited at the three different temperatures has been measured using profilometer Dektak XT (see table 1). the thickness of the LTO thin films was found to be decreased by increasing the annealing temperature, Because the deposited particles enhanced their energy from the substrate and become more active as the resulting of increasing their motions with the temperature, appearing as the thinner thickness of these films [23] which is an inversely proportional relationship (see figure 3).

The roughness of the LTO thin films has been measured by using the Dektak profilometer (see table 2) and it was found to be decreased by increasing the annealing temperature as shown in figure 5. The value of the lowest roughness equal to 12.28 nm for the thinner film with a thickness of 253.46 nm,
while the highest roughness is 14.74 nm for the thicker thin film at 323.05 nm, which is deposited at 700°C and 500°C, respectively.

Table 1. The average thickness of the LTO thin films deposited at different annealing Temperatures.

| Annealing Temperature (°C) | 500   | 600   | 700   |
|----------------------------|-------|-------|-------|
| Thin film thickness (nm)   | 323.05| 289.15| 253.46|

![Image of thickness vs Temperature graph]

Figure 4. The relationship between the thicknesses of the LTO thin films deposited in different temperatures.

Table 2. The roughness value of the LTO thin films deposited in different annealing Temperatures.

| Annealing Temperature (°C) | 500 | 600 | 700 |
|----------------------------|-----|-----|-----|
| Roughness (RMS) nm         | 14.74| 12.43| 12.28|

![Image of roughness vs Temperature graph]

Figure 5. The relationship between the roughnesses of the LTO thin films deposited at different annealing Temperature.

4. Conclusions

In this work, Amorphous LTO thin films were fabricated on Si (100) substrate via pulsed laser deposition technique (PLD) and the impact of the thermal annealing on the thickness and the roughness of deposited films has been studied. The collected results showed that the thickness decreased by increasing the annealing temperature linearly, and the roughness was inversely proportional to the increasing of substrate temperature, so the achieved result confirmed that the thermal annealing of LTO films in a vacuum had a noticeable influence on characterizations of the LTO thin films.

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