11th International Conference on Advanced Materials
ICAM 2009
Rio de Janeiro Brazil
September 20 - 25

Lithium lanthanum titanate thin films prepared through polymeric precursor method

A. H. M. González[(1)*], S. L. Fernandes[(1)], B. Antoniassi[(2)], C. F. O. Graeff[(2)], J. A. Varela[(3)] and E. Longo[(3)]

(1) Department of Chemistry, Faculty of Sciences, UNESP/Bauru. e-mail: alejandra.horten@uol.com.br
(2) Department of Physics, Faculty of Sciences, UNESP/Bauru.
(3) INCTMN/CMDMC, Institute of Chemistry, UNESP/Araraquara.
* Corresponding author.

Abstract – $La_{0.50}Li_{0.50}TiO_3$ thin films were prepared by spin coating method using a polymeric organic solution. The films were deposited on Si (100) substrates and thermally treated from 400°C to 700°C for 3 h in order to study the influence of the thermal treatment temperature on the crystallinity, microstructure, grain size and roughness of the final film. X-ray diffraction (XRD) results showed that the films are polycrystalline and a secondary phase was observed (Fig.1). The thickness of films was determined by scanning electron microscopy (SEM) (Fig.2). The film surface morphology was observed with an atomic force microscope (AFM).

Lithium-conducting materials are of great interest due to their potential use as electrolytes or electrode materials in electrochemical devices such as sensors, rechargeable batteries and miniature supercapacitors [1]. Li-batteries also hold promise for the development of small capacity energy sources, which are in high demand for applications in consumer electronics [2]. The thin film battery is a promising alternative micropower source, which can be miniaturized by a typical thin film process. Discovery of high Li-ion conductivity in lithium lanthanum titanate (LLTO) has generated new interest in this direction [3].

$La_{0.50}Li_{0.50}TiO_3$ precursor solution was prepared by polymeric precursor method. This method is based on metallic citrate polymerization with use of ethylene glycol. LLTO films with 5 layers were deposited on Si (100) substrates by spin coating method at a rotation speed of 5000 rpm for 30 s. Then, the thin films were thermally treated using a two-step heat treatment. A preheating at 300°C for 1 h with heating rate of 3°C /min was used to eliminate water and excess ethylene glycol, and promote the polyesterification process. After that, the films were heated from 400°C to 700°C for 3 h with heating rate of 5°C /min to reach the crystallization stage. The crystallization process of the prepared films was analyzed by X-ray diffraction (XRD). A correlation between the process of phase crystallization and organic fraction elimination was evident. Heating the thin film at 700ºC leads to a crystallization of LLTO phase and a secondary phase (2θ = 18.5° and 43.5°), identified as $Li_2Ti_2O_5$. Average grain size and surface roughness of the LLTO thin films were estimated using a contact mode atomic force microscopy (AFM). The microstructure study revealed that all surfaces are not only crack-free but also appear relatively smooth. At 700°C, the beginning of the crystallization process is observed, which is followed by the growth of grains with size between 0.5 and 0.8 µm. To determine the thickness of LLTO thin films, high-magnification SEM observation was performed. It can be seen that the thickness of the films decreased with increasing the annealing temperature, between 400 and 500°C, due to the densification of films. However, as the temperature increased from 500 to 700°C, both thickness and roughness data of films increased.

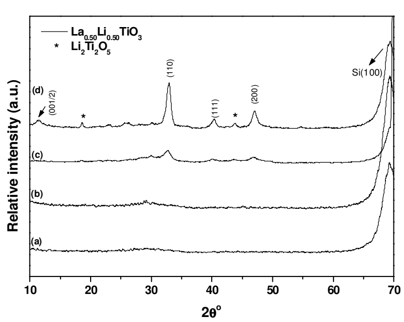

Figure 1: XRD patterns of LLTO thin films deposited on Si (100) and thermally treated at: (a) 400; (b) 500; (c) 600 and (d) 700ºC/3h.

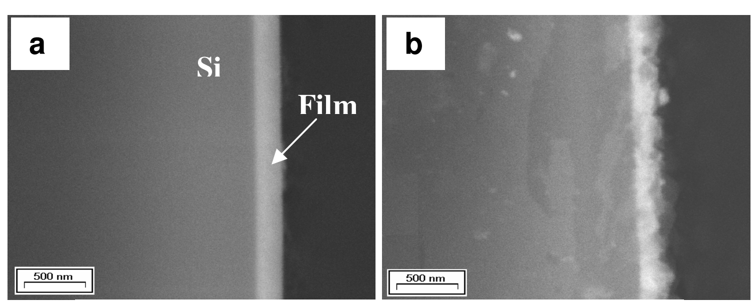

Figure 2: Thickness micrographs obtained by SEM of 5-layered $La_{0.50}Li_{0.50}TiO_3$ thin films deposited on silicon (100), and thermally treated at: (a) 400ºC/3h; (b) 700ºC/3h.

References

[1] A. Belous, O. Yanchevskiy and O. V'yunov. Chemistry Materials, 16 (2004) 407.
[2] J. Kim, H. Lee, K. Lee, S. Lim and S. Lee. Electrochemistry Communications, 5 (2003) 544.
[3] Y. Inaguma, C. Liquan, M. Itoh, T. Nakamura, T. Uchida, H. Ikuta and M. Wakihara. Solid State Communications, 86 (1993) 689.