

Controlling Preferred Orientation of ITO Thin Films by RF-Magnetron Sputtering Method

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Abstract

Sn-doped In_2O_3 (ITO) thin film is one of the materials widely on research not only in the academic fields but also in industrial fields because of their transparency, high conductivity and good adhesion characteristics on substrate. ITO thin films are usually preferred oriented to one of the (222), (400), and (440) planes during crystallization process, which is dependent on processing variables. The preferred orientation affects electrical, optical and etching properties of the films. In this study, thin films of preferred oriented in different orientation were fabricated by controlling processing variables. The crystallization behavior, grain size, surface roughness, transparency and electrical properties of the thin films in different orientation were examined.

1. Introduction

Transparent conducting indium tin oxide (ITO) thin films are extensively used in the electronic and optoelectronic device fabrication technologies. Application of such films in each branch of technology demands certain specific values of conductivity, transmittance, bandgap, etc. [1,2] The wide range of results suggests that the properties depend very much on the preparation conditions like deposition rate, substrate temperature, oxygen partial pressure, etc. The influence of the various experimental parameters for different methods has been analyzed.[3,4]

Preferred orientation of a film can be understood based on energy considerations: the surface energy, the epitaxial bonding energy, and the elastic energy in the film. If one considers structural property, the available reports on the reactive thermal deposition technique reveals that the deposited films were

oriented with (222) or (400) predominant planes.[5] Spray pyrolysis deposition yielded films with (400) predominant planes.[6] However, there is no clear report on the change in predominant planes of the films prepared by the same method.

In this study, preferred orientation of ITO thin films was controlled by changing various sputtering conditions. Surface characteristics and resistivity of the films were also analyzed.

2. Experimental procedure

The ITO target was prepared by the general solid-state reaction method of oxides. High purity nanocrystalline In_2O_3 and SnO_2 powders were used as the raw materials. A mixed powder corresponding to the $\text{In}_2\text{O}_3/\text{SnO}_2$ weight ratio of 9/1 was prepared. The powder was formed into 150 mm disk by cold isostatic pressing (CIP) at 300 MPa, then sintered at 1550 °C for 10 h in oxygen atmosphere. The apparent density of the target was 96% and the dimension of the target was adjusted to 76mm in radius and 5mm in thickness. The substrate of SiO_2 coated Corning 1737 glass was cleaned in acetone, methyl alcohol, and deionized water in sequence by ultra sonic cleaner, then dried with nitrogen gas. Prior to the deposition, the chamber was evacuated to a background pressure of 1.0×10^{-6} torr initially, and the working pressure during film deposition was varied from 5 to 40 mTorr by utilizing Ar and O_2 gas, and the O_2 content was varied from 0 to 60%. The distance between the target and the substrate was 5 cm, and RF power was

varied from 25 to 100 W. The deposition time was also varied from 5 to 80 min. The deposition process was carried out at room temperature, i.e. the substrate was not heated during and after the film deposition. Film crystalline structure was investigated by X-ray diffractometer using Cu-K α radiation. Wavelength dependent absorption and transmission of the ITO films were measured by a UV-VIS-NIR spectrophotometer over the wavelength range of 200-800 nm. Sheet resistance of the films was measured by a 4-point probe Sheet Resistance/Resistivity Measurement System and the surface morphology of the films was observed and analyzed with a scanning electron microscope and a scanning probe microscope, respectively.

3. Result and discussion

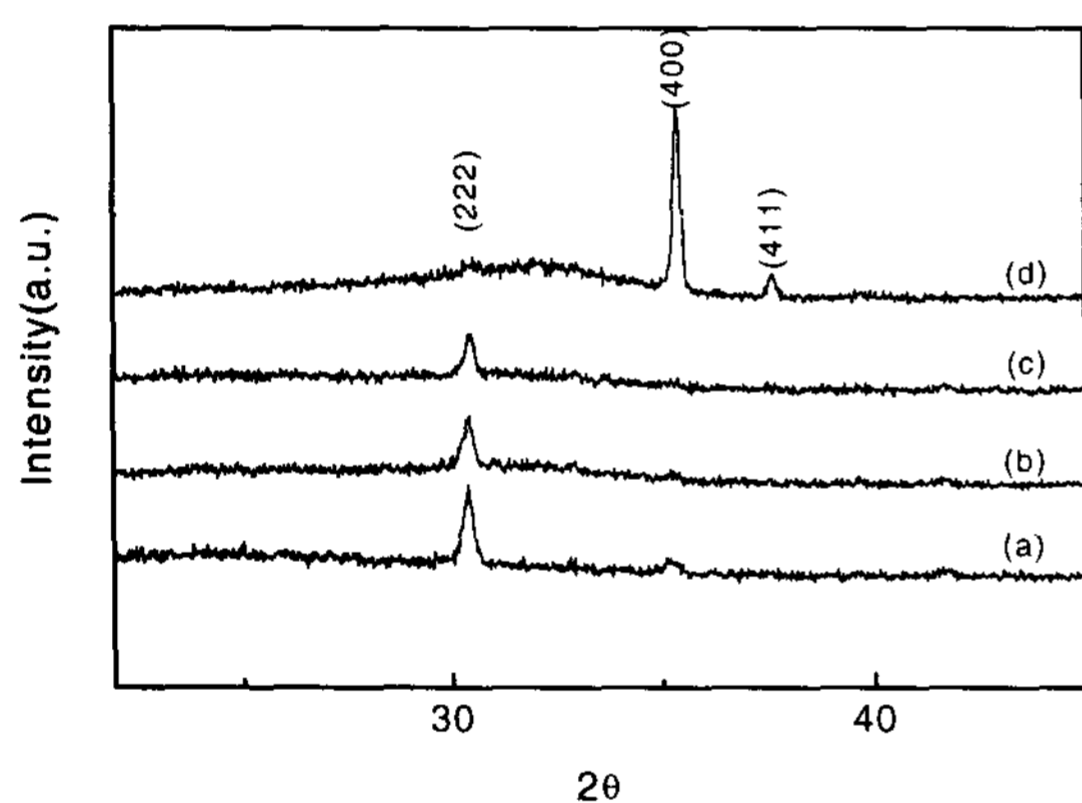


Fig. 1. X-ray diffraction patterns of ITO thin films as a function of oxygen flow rate of Ar:O₂ (a) 2:3, (b) 3:2, (c) 4:1 and (d) 5:0

Fig. 1 shows the X-ray diffraction patterns of thin films deposited as a function of oxygen volume fraction under the conditions of working pressure 5 mTorr, RF power 50W, deposition time 80 min. As the volume fraction of oxygen increased the intensity of (222) predominant plane increased.[7] Fig. 2 shows the result of X-ray diffraction patterns of thin films when the oxygen volume fraction, working pressure and RF power are fixed to Ar:O₂=2:3, 10 mTorr, 25 W, respectively, and deposition time varies. When the deposition time

increased, the Crystallization of the films was activated and the preferred orientation of (222) plane outstandingly increased at the same time.

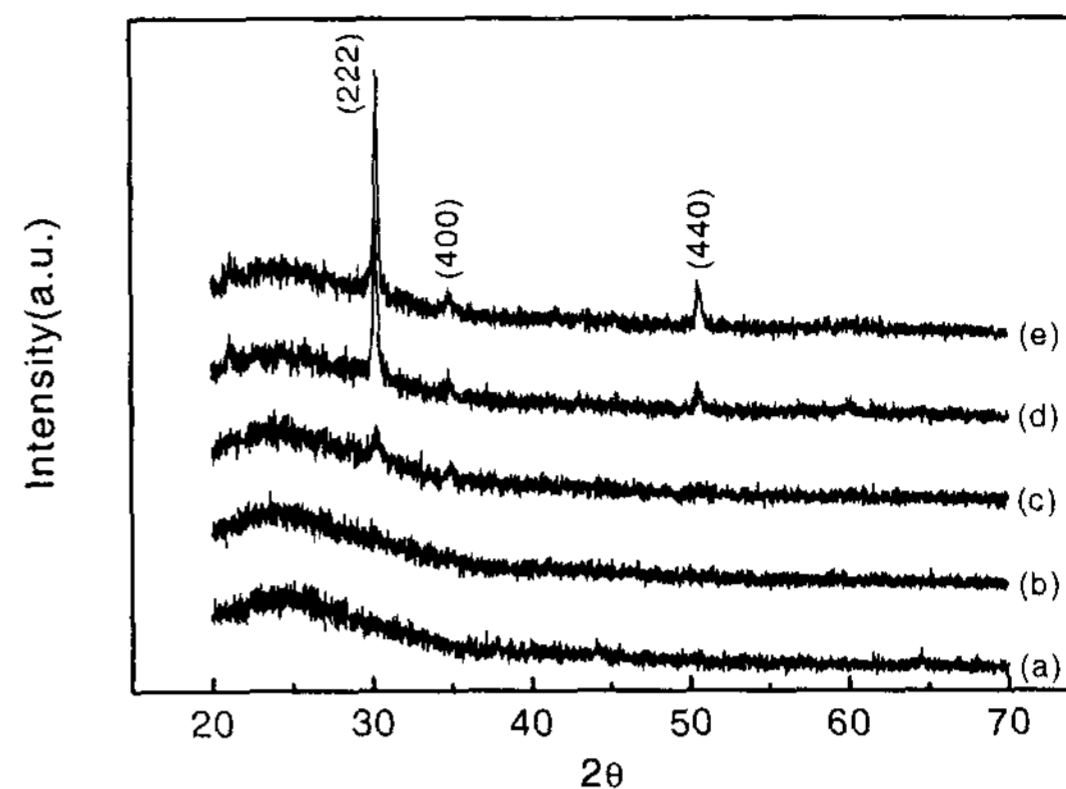


Fig.2. X-ray diffraction patterns of ITO thin films as a function of deposition time (a) 5, (b)10, (c) 20, (d) 40 and (e) 80 min.

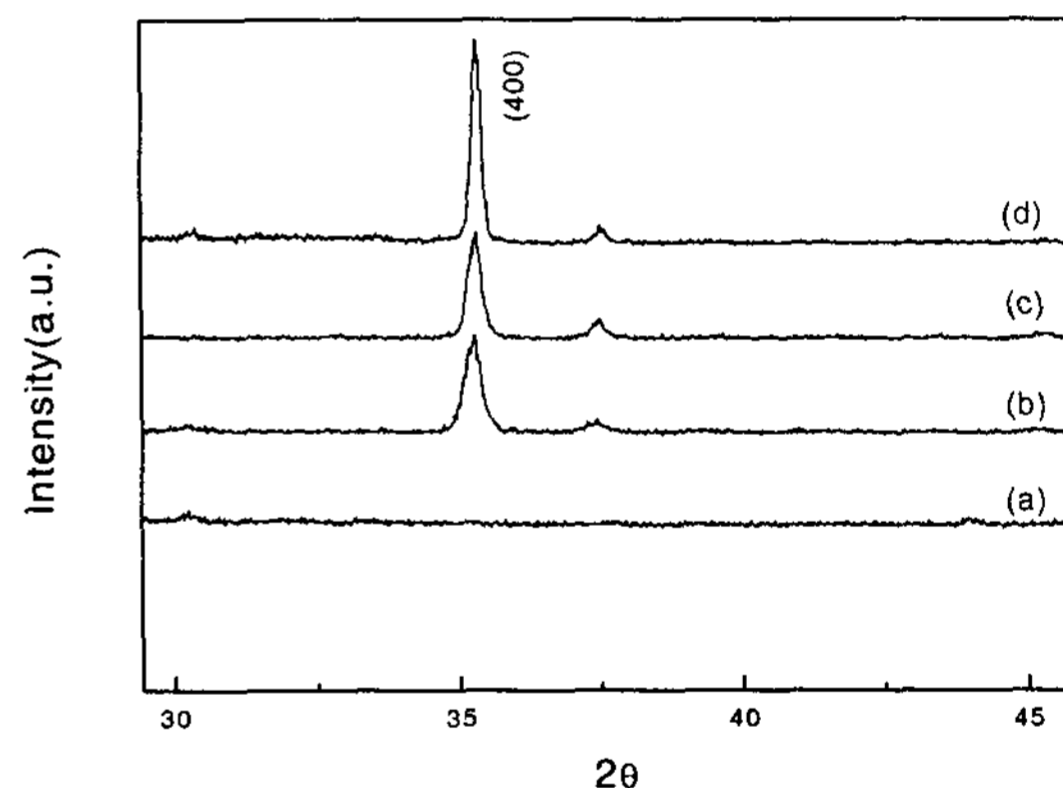


Fig. 3. X-ray diffraction patterns of ITO thin films as a function of RF power (a) 25, (b) 50, (c) 75 and (d) 100 W.

Fig. 3 shows the X-ray diffraction patterns of thin films deposited as a function of RF power under the condition of working pressure 5 mTorr, oxygen volume fraction 0 %, deposition time 80 min. As the RF power increased predominant peak along (400) plane increased.

Fig. 4 shows the result of X-ray diffraction patterns of thin films when the RF power is fixed at 50W and deposition time varies.

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The preferred orientation of (440) plane increased when mean free path was shortened by increasing working pressure in the condition of (400) preferred orientation.

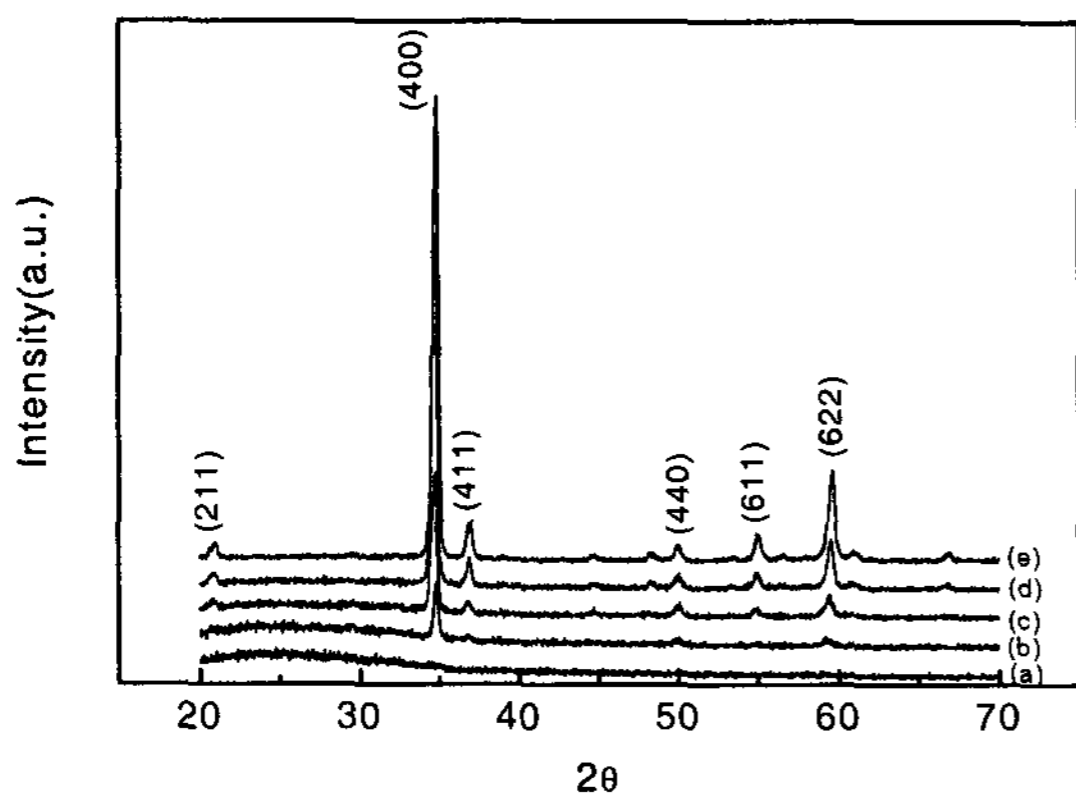


Fig. 4. X-ray diffraction patterns of ITO thin films as a function of deposition time (a) 5, (b)10, (c) 20, (d) 40 and (e) 80 min.

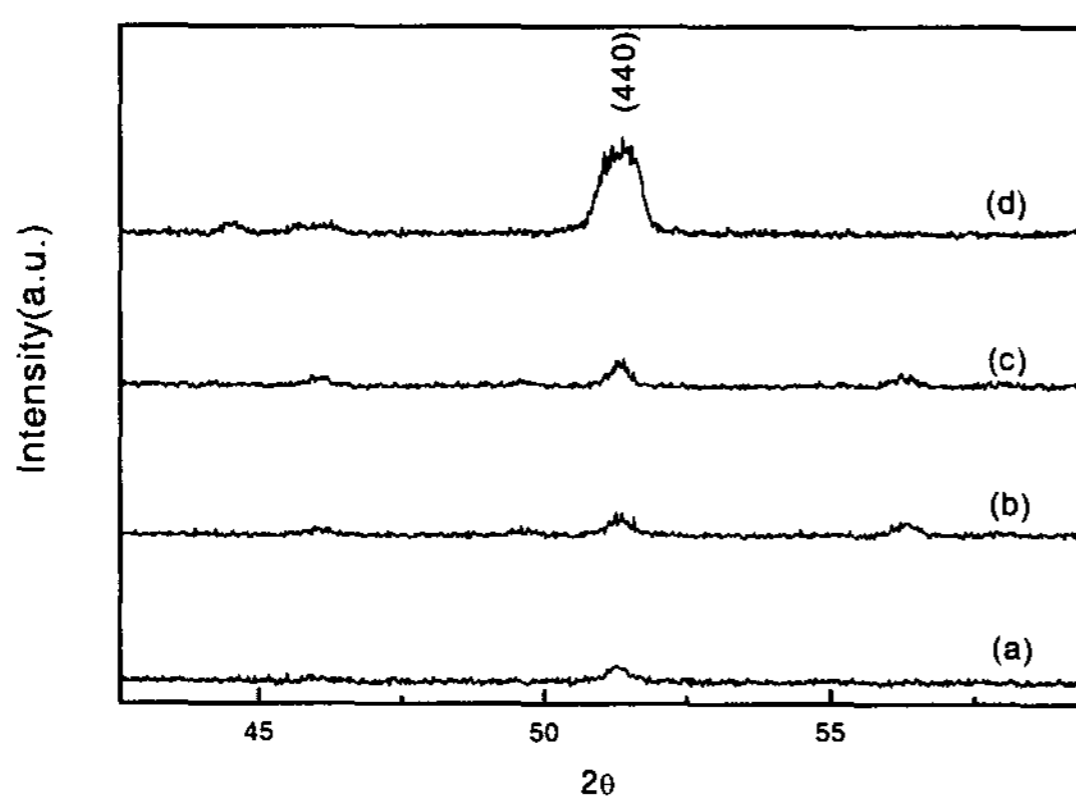


Fig. 5. X-ray diffraction patterns of ITO thin films as a function of working pressure of (a) 5, (b) 10, (c) 20 and (d) 40 mTorr.

Fig. 5 shows the X-ray diffraction patterns of thin films deposited as a function of working pressure under the condition of oxygen volume fraction 0 %, RF power 50W, deposition time 80 min. As the working pressure increased preferred orientation of (440) plane increased.

Fig. 6 represents the X-ray diffraction profiles of films when the working pressure is fixed to 40 mTorr and deposition time changed under the same condition with that of Fig.5. As the deposition time

increased, preferred orientation of (440) plane greatly increased

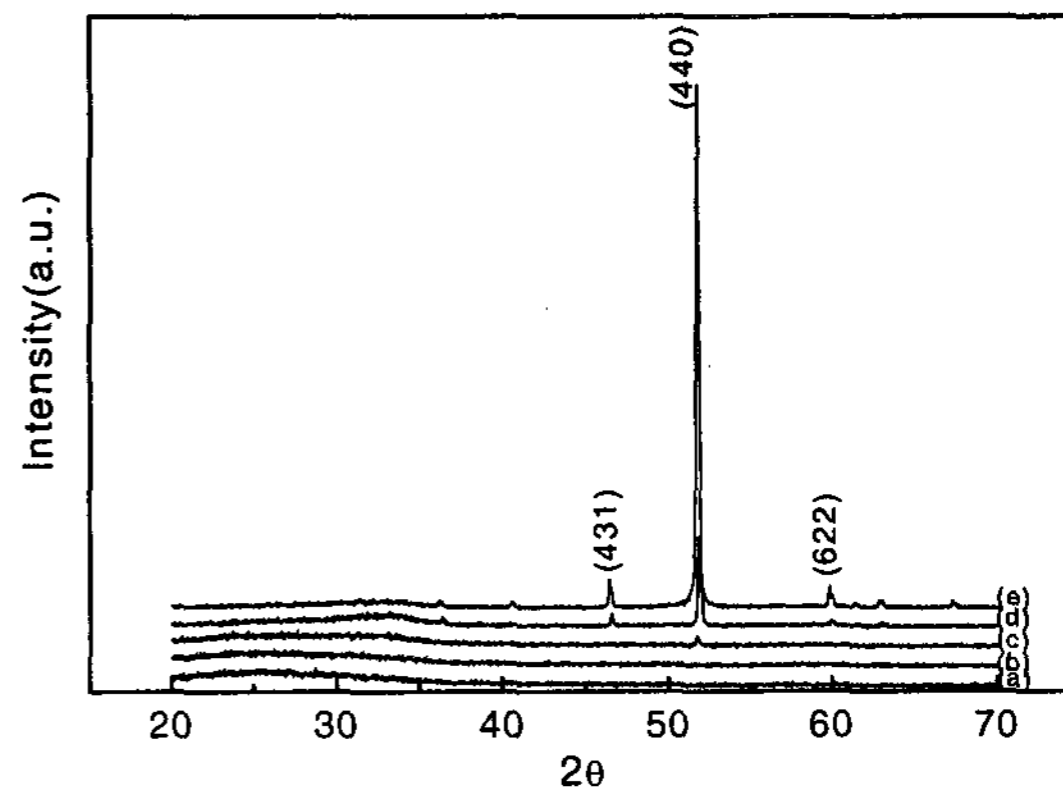


Fig. 6. X-ray diffraction patterns of ITO thin films as a function of deposition time (a) 5, (b)10, (c) 20, (d) 40 and (e) 80 min.

The surface of thin films was analyzed by SEM and SPM. Even though the photographs of microstructures are not presented here, the surface of (222) preferred orientation was very smooth with a mean grain size below 10nm, and the thickness of the film was relatively thin comparing with other films with different preferred orientation. This seems to be caused by inhibited film growing due to the lower RF power which results in less number of sputtered particles. The increased volume fraction of O₂ reduces the relative volume fraction of Ar, which makes the deposition less effective. On the other hand, the films of (400) preferred orientation was thick comparatively and has rough surface with the mean grain size of 10~40 nm and their cross sectional view was a columnar structure. This result was caused by lower working pressure and higher RF power that makes the deposition effective. The film of (440) preferred orientation has less surface roughness than the surface of (400) and has the mean grain size of 100~200nm. Small subgrains of 5~20 nm were observed in each grain. This was caused by the increase of

working pressure when compared to the working pressure of (400) plane, that leads to the increase of mean free path and energy reduction of sputtered particles.

Thin films with (222) preferred orientation have high bandgap energy and low electrical conductivity whereas the films with (400) preferred orientation showed good electrical conductivity and worse optical characteristics.

Table 1. Characteristics of ITO thin films with different

Preferred orientation	(222)	(400)	(440)
Roughness(nm) (R_{rms})	0.21	5.64	3.61
Bandgap (eV)	4.21	3.45	3.81
Resistivity($\times 10^{-3} \Omega \cdot \text{cm}$)	15.4	1.4	3.0

4. Conclusion

In the RF sputtering method, various processing parameters were varied and thin ITO films with different preferred orientation are produced. The surface roughness, optical bandgap and resistivity were influenced on the preferred orientation of the films. In case of (222) oriented film, surface was very smooth, while bandgap and resistivity revealed the largest values comparing to the other films. In case of (400) oriented film, it showed the best conductivity but worst surface roughness among the films with different preferred orientation.

5. Acknowledgement

This work was supported by grant No. (R12-2002-055-01003-0) from the Basic Research Program of the Korea Science & Engineering Foundation.

References

- [1] K. L. Chopra, S Major and D. K. Pandya, Thin Solid Films, 102, p. 1 (1983)
- [2] Z. Ovadyahu and Hn Wiesmann, J. Appl. Phys., 52, p. 5865 (1981)
- [3] R. N. Joshi, V. P. Singh, J. C. McClure, Thin Solid Films., 257, p.32 (1995)
- [4] E. Terzini, G, Nobile, S. Loreti, C. Minarini, T. Polichetti and P. Thilakan" 38, p. 3448 (1999)

- [5] P. Thilakan, S. Kalainathan, J. Kumar and P. Ramasamy, J. Electronic Materials, 20, p. 719 (1995)
- [6] V. Vasu and A. Subramahmanyam, Thin Solid Films, 193-194, p. 696 (1994)
- [7] C. G. Choi, K. S. No, W. J. Lee, H. G. Kim, S. O. Jung, W. J. Lee, W. S. Kim, S. J. Kim and C. Yoon Thin Solid Films, 258, p. 274 (1995)