THE INFLUENCE OF GALLIUM (Ga) CONTENT ON MORPHOLOGICAL, STRUCTURAL AND OPTICAL PROPERTIES OF INDIUM TIN OXIDE THIN FILMS

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Abstract. *This paper reports on preparation of ITO thin films with different concentrations of Ga by spay pyrolysis method on Si (100) substrates. The morphology of the prepared films was studied using scanning electron microscope (SEM), and the quantitative chemical composition was determined using energy dispersive X-ray spectroscopy (EDX). X-ray diffraction (XRD) measurements were carried out on Bruker D8 Advance X-Ray Diffractometer with CoKα radiation (λ=0.1789 nm) in the 2Θ region of 20° - 90°. The optical properties of ITO:Ga thin films were measured using a Jasco V-670 spectrometer at room temperature (300 K).*

Keywords: *SEM, EDX, ITO, X-ray spectroscopy, thin films.*

Introduction

Currently, great attention is paid to the development of optoelectronic devices for the ultraviolet (UV) region of the spectrum. One of the promising and intensively explored materials is indium tin oxide (ITO). Particularly, ITO thin films are typically used in gas sensors [1-2], semiconductor devices [3], solar cells [4-6] and also as UV radiation detectors or light emitters [7-9], due to its high optical transparency, thermal and chemical stability. ITO is easily obtained by various methods such as RF magnetron sputtering [10,11], electron beam deposition [12], excimer laser evaporation [13], pulse laser deposition PLD [14-15], thermal evaporation technique [16], laser-vacuum deposition [17], and microwave heating [18]. However, the spray pyrolysis used in this study is one of the most cost efficient and fast methods [19]. Addition of Ga to this material system can be applied for changing the band gap, thus shifting the emission spectrum or sensitivity spectral range to shorter wavelengths. This combination ensures the possibility of obtaining better luminescent, photoelectric and optical properties in a fairly wide range from 3.5 eV (ITO) to 4.9 eV (Ga2O3) by adjusting the Ga content.

Sample preparation of ITO:Ga

Thin films of ITO:Ga were deposited at 460°C on n-Si (100) substrate by spray pyrolysis method. First of all, the Si substrates were chemically degreased in a chemical solution with a volume fraction of H_2O:NH_3:H_2O_2 equal to 8:1:1, at a temperature of 80 °C for 10 minutes. It was then kept in hydrofluoric acid for 2 minutes, followed by washing in distilled water and immediately starting the deposition process. Each individual solution of indium chloride ($InCl_3$), tin chloride ($SnCl_4$) and gallium nitrate ($Ga(NO_3)_3$) with molar mass (0.5M) were dissolved in ethanol (C_2H_5OH) and mixed in an ultrasonic bath for 30 minutes at a temperature of 50 °C for the aerosol deposition process. Mixtures of ITO and Gallium were taken in proportions as follows: ITO, $ITO_{0.8}Ga_{0.2}$, $ITO_{0.7}Ga_{0.3}$ and $ITO_{0.6}Ga_{0.4}$. The deposition time for each thin film was 10 minutes. The solution was sprayed by means of a sprayer at an oxygen flow of 0.1 atm in excess of normal atmospheric pressure.

Results and discussions

Morphological characterization and analysis of the chemical composition of the thin films were performed with a scanning electron microscope LEO-ZEISS Gemini 1530, with an energy dispersion X-ray analysis unit (EDX). The morphology of ITO:Ga thin films was found to change with increasing the Ga content from x = 0.00 to x = 0.40, as observed from SEM images presented in Fig. 1.

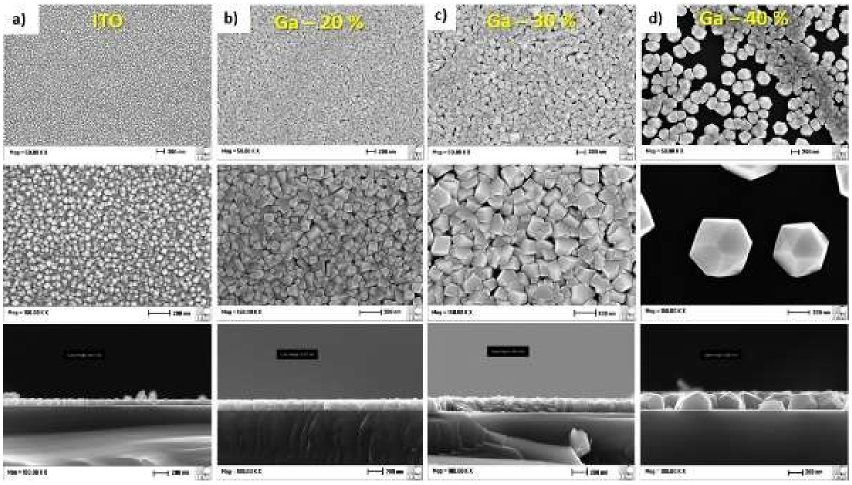


Figure 1. SEM images of ITO:Ga thin films obtained by spray pyrolysis method

As can be seen from images, a higher concentration of Ga leads to an increase in nanoparticles size, contributing to reduction of their number on the surface of the substrate. At the same time, the thickness of the thin films increases from 90 nm (ITO) to 260 nm (ITO:Ga), keeping the same deposition time of 10 minutes. The thickness of the deposited films is related to the concentration of Ga, as presented in Fig. 2.

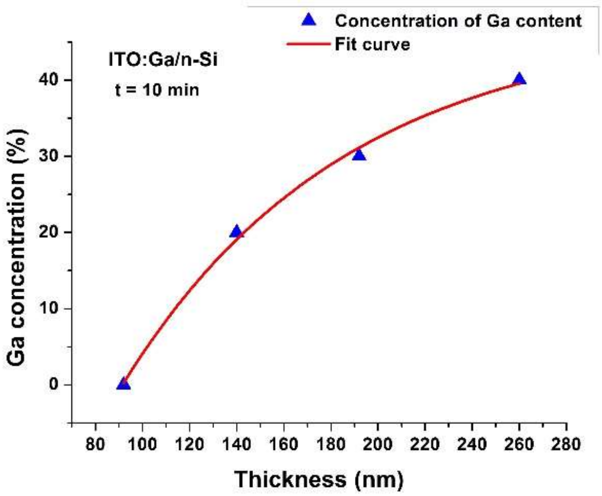
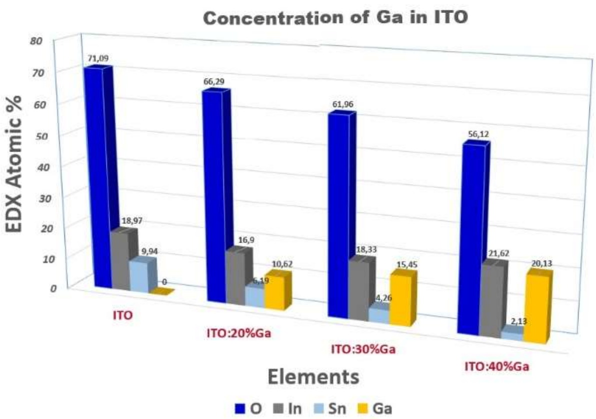


Figure 2. Relation between the thin films thickness and the concentration of Ga

The chemical composition analysis of ITO:Ga thin films obtaining by spray pyrolysis method is shown in Fig. 3. One can see from this analysis, that the content of Ga in the deposited films is two times lower that the expected one from the precursor solution.



Fugure 3. EDX analysis of ITO:Ga thin films obtained by spray pyrolysis method

X-ray diffraction (XRD) measurements (Fig. 4) were carried out on a Bruker D8 Advance X-Ray Diffractometer with CoKα radiation (λ=0.1789 nm) in the 2Θ region of 20° - 90°. It should be noted that the reflexes do not change their positions, but just change their intensity. The 110, 431, 220, and 543 reflexes disappear at 20 % Ga content. However, they appear again at a Ga content of 30 %.

This observation can be explained by changing the structural quality of ITO thin films. The intensity of reflexes decreases again at Ga content of 40 %, which can be due to the reduction of nanoparticles number on the surface of the substrate, thus creating nano-islands. The high intensity peaks corresponding to planes (400), (420) and (332) were observed as predominant peaks for all the thin films obtained.

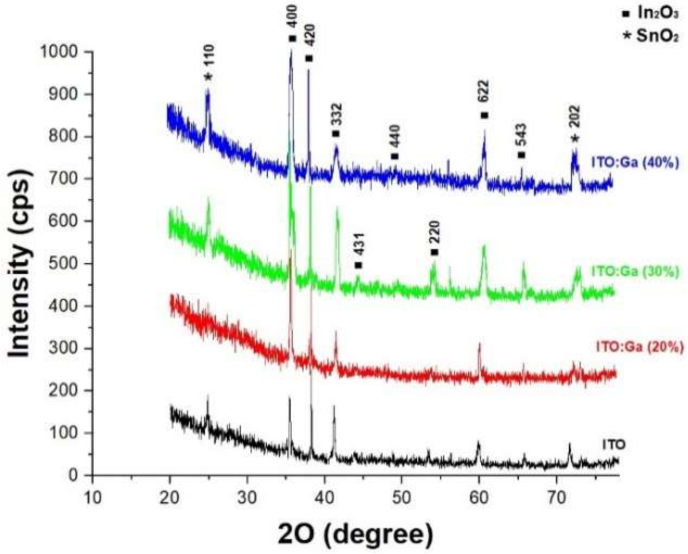


Figure 4. X-ray diffraction patterns of ITO:Ga thin films

The optical properties of ITO:Ga thin films were measured at room temperature (300 K) using a Jasco V-670 spectrometer. The band gap was determined from the Tauc plot of the absorption coefficient, and the results are shown in Fig. 5. From this graph one can see that the band gap of thin films can be adjusted by changing the Ga content, thus increasing it from 3.55 eV for ITO to 4.1 eV for 40% Ga concentration.

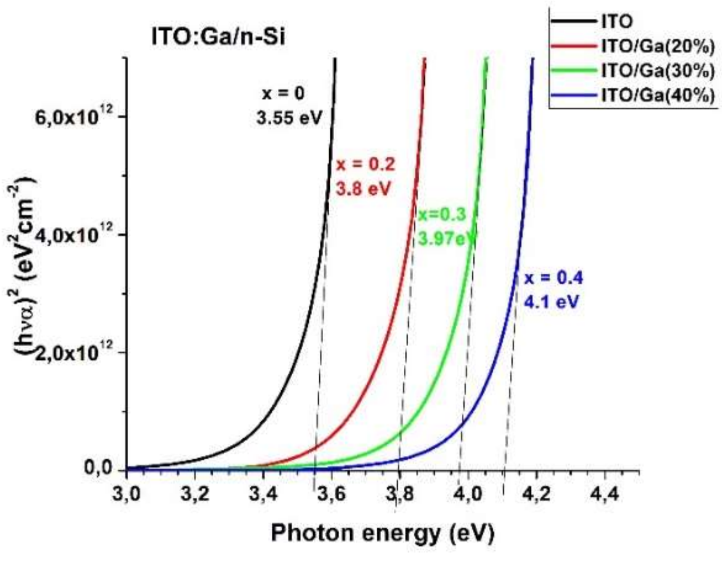


Figure 5. Tauc plot of optical absorption spectra measured at room temperature for ITO:Ga films deposited by spray pyrolysis methods on Si substrates

Conclusions

The results of this study demonstrate the influence of Ga content on the morphological, structural and optical properties of ITO thin films with x value from 0.0 to 0.4 prepared by spray pyrolysis deposition on Si (100) substrates. It has been established that the increase of Ga concentration leads to the increase of nanoparticles size, as well as the thickness of thin films from 90 nm for ITO to 260 nm for films with 40% Ga content, with the same duration of the deposition process of 10 minutes. As the concentration of Ga increases, the band gap of the material also increases, it reaching a value of 4.1 eV for 40 % of Ga content.

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