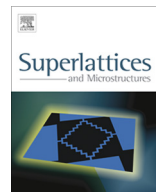




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Synthesis and characterization of Mo doped SnO₂ thin films with spray pyrolysis



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ABSTRACT

Tin oxide thin films doped with different Mo content were successfully grown by spray pyrolysis and they were characterized as a function of Mo content, changed from 0 to 3.5 at.% with 0.5 at.% step. The XRD studies showed that the films had SnO₂ cassiterite structure with (2 1 1) preferential orientation and the best crystal properties was observed for 2 at.% Mo doped sample. The SEM images indicated the films were made up of nanosized grains and it was observed pyramidal, polyhedron shaped grains on the deposited films' surfaces. From electrical and optical studies, although 2 at.% Mo doped SnO₂ film exhibited the lowest sheet resistance (39.81 Ω) and the highest IR reflective (81.77%), 1 at.% Mo doped film has the highest optical band gap (4.011 eV). The lowest Urbach energy (293 meV) and the highest figure of merit ($1.80 \times 10^{-3} \Omega^{-1}$) values were observed for 0.5 at.% Mo doped sample between all films. The results found in present study showed that Mo doped SnO₂ thin films is a good candidate for solar cells, IR coating and other optoelectronic and technological applications.

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1. Introduction

Recently, there is a growing effort on wide band gap oxide semiconductors owing to their excellent optical, electrical, chemical, mechanical properties [1], abundance in nature, non-toxicity, low-cost

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and easy fabrication [2,3]. One of these oxide semiconductors, tin oxide-TO-SnO₂, has wide range of application areas such as gas sensing devices, transparent conducting electrodes, liquid crystal displays [4,5], window layers, heat reflectors in solar cells [6], optoelectronic devices and photo-catalysts [7]. In order to increase efficiency of SnO₂ in technological applications, many studies on fabrication of SnO₂ thin films have been made up to the present with several thin film deposition technique. The spray pyrolysis is one of the most effective methods at growing of SnO₂ thin films. It has many advantages such as inexpensive and simple experimental setup [8], easy control of chemical specimens, reproducibility and uniform and large area coating [9].

In spray pyrolysis process of SnO₂, the features of SnO₂ depend on the spraying parameters; substrate temperature, spray duration and flow rate of carrier gas, nozzle-substrate distance, ambient atmosphere, spray solution content (solvent type, doping element, chemical salt type, etc.), substrate type, etc. [10–13]. The doping with foreign atoms greatly affects the properties of SnO₂ thin films. The doping of SnO₂ structure can be achieved by replacing X^{m+} cations and Y^{n-} anions with Sn^{4+} and O^{2-} ions [14]. If $m > 4$ and $n < 2$, X and Y ions give extra free electrons to SnO₂ structure and they cause to increasing electrical conductivity and optical transparency and reflectance. The doping elements can also improve microstructural, morphological and mechanical features of SnO₂.

In literature, numerous works have been carried out on doping cation Sb and anion F into SnO₂ [15–21], and it found that Sb and F elements improved some properties of tin oxide structures. However, nowadays, SnO₂ structure have been doping with Nb, V, Mo, W elements as alternative cations and their effect on SnO₂ have been investigated. Molybdenum (Mo) has many oxidation states up to Mo⁶⁺ like tungsten (W) [22], and it is expected that electrical conductivity, optical transparency and reflectance of SnO₂ increase with Mo doping because of giving two extra free electrons to SnO₂ lattice. A great number of detailed studies on spray deposited Mo doped SnO₂ thin films should be made in order to investigate Mo doping effect on properties of SnO₂, because, in earlier studies, there are a few studies have been made on Mo doped SnO₂ thin films [23–33], and to our best knowledge, there is no study about Mo doped SnO₂ thin films with spray pyrolysis. Thus, we aimed to deposit Mo doped SnO₂ thin films on glass substrates with spray pyrolysis and to investigate Mo doping effect on microstructural, morphological, optical and electrical properties.

2. Experimental details

SnO₂ thin films doped with different Mo content from 0 at.% to 4 at.% with step of 0.5 at.% were prepared by an economic spray pyrolysis method. The essential amounts of SnCl₂·2H₂O and MoCl₆ salts were dissolved in propane-2-ol solvent contained of a few drops of HCl in order to prepare undoped (TO), 0.5 at.% Mo doped (MTO-0.5), 1 at.% Mo doped (MTO-1), 1.5 at.% Mo doped (MTO-1.5), 2 at.% Mo doped (MTO-2), 2.5 at.% Mo doped (MTO-2.5), 3 at.% Mo doped (MTO-3), 3.5 at.% Mo doped (MTO-3.5), 4 at.% Mo doped (MTO-4) spray solutions. After these solutions were prepared, they were stirred for two hours for obtaining more homogenous solution then they were sprayed onto preheated hot glass substrates, which were cleaned with acetone, methanol and deionized water in ten-minute periods. The substrate temperature (500 °C), nozzle-substrate distance (40 cm), the flow rate of filtered carrier air (1.5 ml/min.), spray solutions quantity (25 ml) were kept constant. After the substrates were deposited, they were characterized with Scanning Electron Microscope (FEI inspect S50 SEM), X-ray Diffraction (XRD) using Rigaku Miniflex II Diffractometer, four point electrical measurements, UV–VIS spectrophotometer (Perkin-Elmer, Lambda 35). All measurement were taken at room temperature.

3. Results and discussion

3.1. Microstructural characterization

The microstructural characterization of the films are made with X-ray diffraction spectra (XRD). Fig. 1 shows the XRD curves of undoped and Mo doped SnO₂ thin films. It is seen that all peaks belong to SnO₂ tetragonal cassiterite structure (JCPDS 41-1445). SnO, Sn₂O₃ and any peak belonging to Mo are

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