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INFLUENCE OF SUBSTRATE TEMPERATURE ON STRUCTURAL AND MORPHOLOGICAL PROPERTIES OF SnO_2 NANOSTRUCTURED THIN FILMS

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ABSTRACT

SnO_2 nanostructures thin films with thickness of 500 nm were prepared by electron beam-physical vapor deposition on glass substrate at temperature of 300, 373, 443, and 583 K. Structural and morphological properties of these nanostructured thin films were studied by Scanning and Transmission Electron Microscopy (SEM, TEM) and Atomic Force Microscope (AFM) methods. The changes in structural and morphological properties are found at different temperatures. Increase temperature causes important change of the structural and morphological properties. The sample prepared at 300 K has crystalline structure and the sample prepared at 583 K has amorphous structure. Roughness parameters have low values at 300, 373, 443 K as opposed to the values obtained at 583 K. This different behavior may be due to the amorphous structure of the sample that was observed in the TEM analysis.

KEYWORDS: electron beam-physical vapor deposition, structural, morphological properties, TEM, SEM, AFM

1. Introduction

Nanostructures of SnO_2 thin films have been employed in a wide range of applications including solid state gas sensors, liquid crystal displays, transparent conducting electrodes, infrared reflectors, plasma display panels (PDPs), transistors, etc. [1–5].

Among such semiconductors, SnO_2 nanostructured thin films seem to be a good candidate for the solar cell applications (e.g., as a window material) because of there wide band gap (3.6 eV) at room temperature [6–10]. In addition, SnO_2 is useful as a hard film material for applications requiring high refractive and reflective properties. SnO_2 thin films have been prepared by various techniques such as RF-magnetron sputtering [1, 11, 12], electron beam evaporation [13], sol–gel coating [14], chemical vapor deposition [15], etc. Various characteristics of these SnO2 films have been observed to change by changing the preparation technique and environment.

Of all the methods, electron beam-physical vapor deposition (EB-PVD) is much preferred for many desirable characteristics such as flexible deposition parameters, low contamination relatively controlled composition, high deposition rates, dense coatings, tailored microstructure [16, 17].

SnO_2 films produced by the EB-PVD process usually have a good surface finish and a uniform microstructure. The versatility of the EB-PVD process is very wide and friendly to environment and new films and materials continue to be developed. In the present study were prepared SnO_2 nanostructured thin films with 500 nm thickness onto glass substrate by EB-PVD at various substrate temperatures.

The influence of substrate temperature on structural and morphological properties of nanostructured SnO_2 thin films were studied using Scanning and Transmission Electron Microscopy (SEM, TEM) and Atomic Force Microscope (AFM).

2. Experimental part

SnO_2 thin films were deposited using electron beam evaporation of SnO_2 powder (Sigma Aldrich, purity 99.99%) as a starting material onto high-quality microscope glass substrates using tungsten crucibles. The substrates were chemically cleaned and by glow discharge 30 mA for 6 minutes. The temperature of the glass substrates during the electron beam evaporation was maintained at different values of 300 K, 373 K, 443K, 583 K in order to study the temperature's influence on the properties of the obtained films. The system was pumped to a base pressure of less than 10^{-3} Pa before deposition and O_2 was injected into the chamber during evaporation at a partial pressure below 10^{-4} Pa. The substrate was set at a distance of 56 cm from the source and rotated at 20 rpm during deposition to obtain uniform and homogeneous films. The deposition parameters were optimized to reduce the film roughness. Thickness of film and rate of its deposition were controlled with the help of an in situ quartz crystal thickness monitor. Thickness of the film was about 500 nm and the deposition rate was set as 20 Å/sec. The TEM investigation was performed on a Philips CM-120 electronic microscope with an accelerating voltage of 120 kV. The surface morphology for SnO_2 nanostructured thin films was studied by Atomic Force Microscope (AFM, model: Solver PRO-M) and scanning electronic microscope (SEM, model: FEI Nova NanoSEM 630).

3. Results and discussion

Selected area electron diffraction (SAED) patterns were taken from a selected area of the SnO_2 thin films. In Fig.1 is shown the SAED pattern of the SnO_2 thin film deposited at substrate temperature of 300 K. The seven intense diffraction rings shown are corresponding to diffraction from (101), (111), (210), (211), (310), (320) and (321) crystallographic planes. The SnO_2 thin film deposited at 300 K exhibits the polycrystalline structure and corresponds to a tetragonal rutile structure.

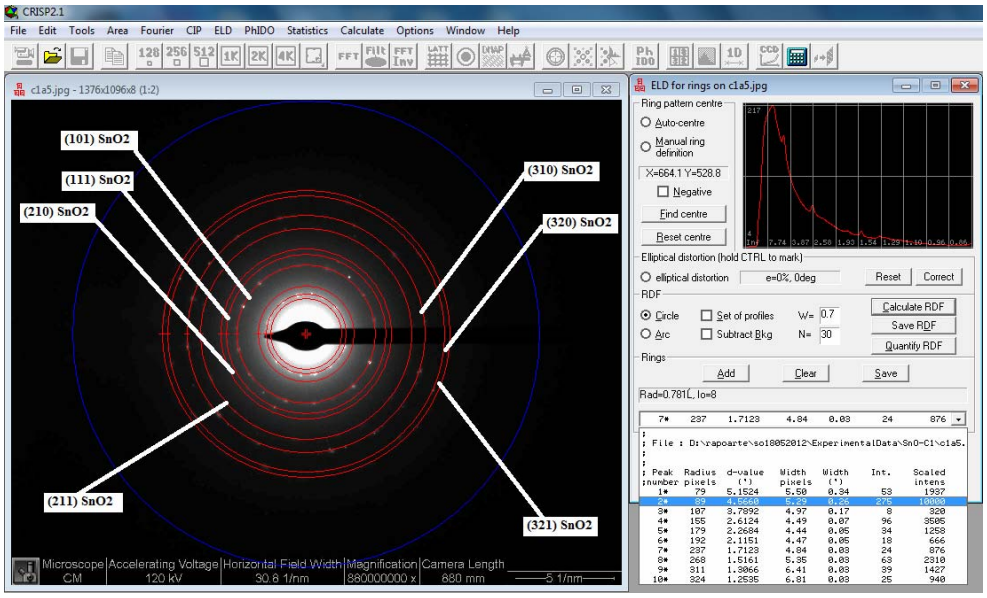


***Fig. 1**. SAED pattern of the SnO_2 thin film deposited at substrate temperature of 300 K*

Fig. 2 shows bright-field transmission electron microscopy (BF-TEM) image recorded from plan view specimen, showing that SnO_2 thin films exhibit a grainy structure which consists of many small grains of relatively uniform size forming a morphologically homogeneous structure.

The size distribution of the crystallites, are plotted in Fig. 3. The distributions of grain sizes, as measured from BF-TEM images, were fitted to the lognormal curves [18, 19]. The mean grain size was found to be around of 9.48nm. Table 1 shows the average particle size varies with the substrate temperature. Note that the maximum average size of 40.78nm of nanoparticles was obtained at temperatures of 373K.

At a temperature of 300K, the average particle size is the smallest value, i.e. 9.48nm. At temperatures of 443K and 583K, nanoparticles average size decreases to 17.16nm and 13.18nm, respectively.

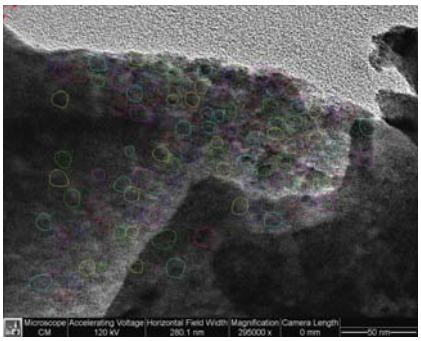


Fig. 2. *BF-TEM images of the SnO_2 thin film*

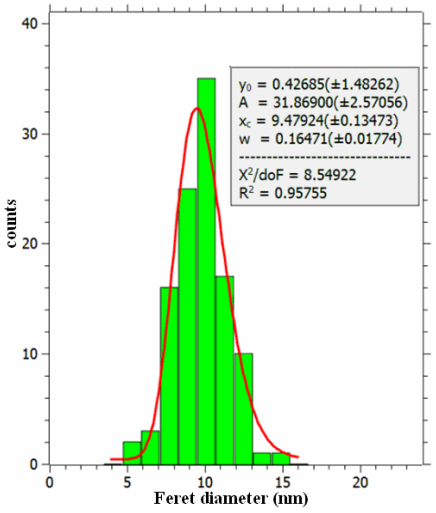


Fig. 3. *The distribution curve of the diameters of the crystalline grains of SnO2 thin films*

Table 1. *Some characteristics of SnO_2 thin films*

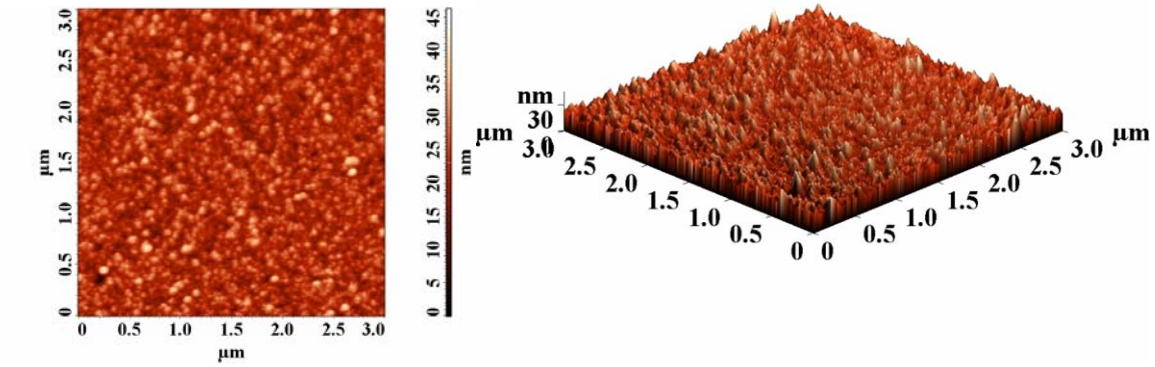
Substrate temperature [K]	**Film thickness** [nm]	**Mean grain size**	**Grain shape**
300	500	9.48	spherical
373	500	40.78	spherical
443	500	17.16	spherical
583	500	13.18	spherical

We find that spherical nanoparticles retain all four temperatures [20, 21].

The structure of the thin films is crystalline at a substrate temperature of 300 K, and it is noticed become amorphous at 583 K.

Another method of investigating the structure of SnO_2 nanostructured thin films is atomic force microscopy (AFM). By this method it has been determined the roughness of the samples studied. Fig. 3 shows 2D and 3D images of SnO_2 samples obtained at temperatures of 300 K (a) 373 K (b) 443 K(c) and 583 K(d) and the 500nm thickness.

Table 2 shows roughness parameters (average roughness, Sa, root square average, Sq) at samples with thickness of 500 nm obtained at temperatures of 300, 373, 443, and 583 K.



a)

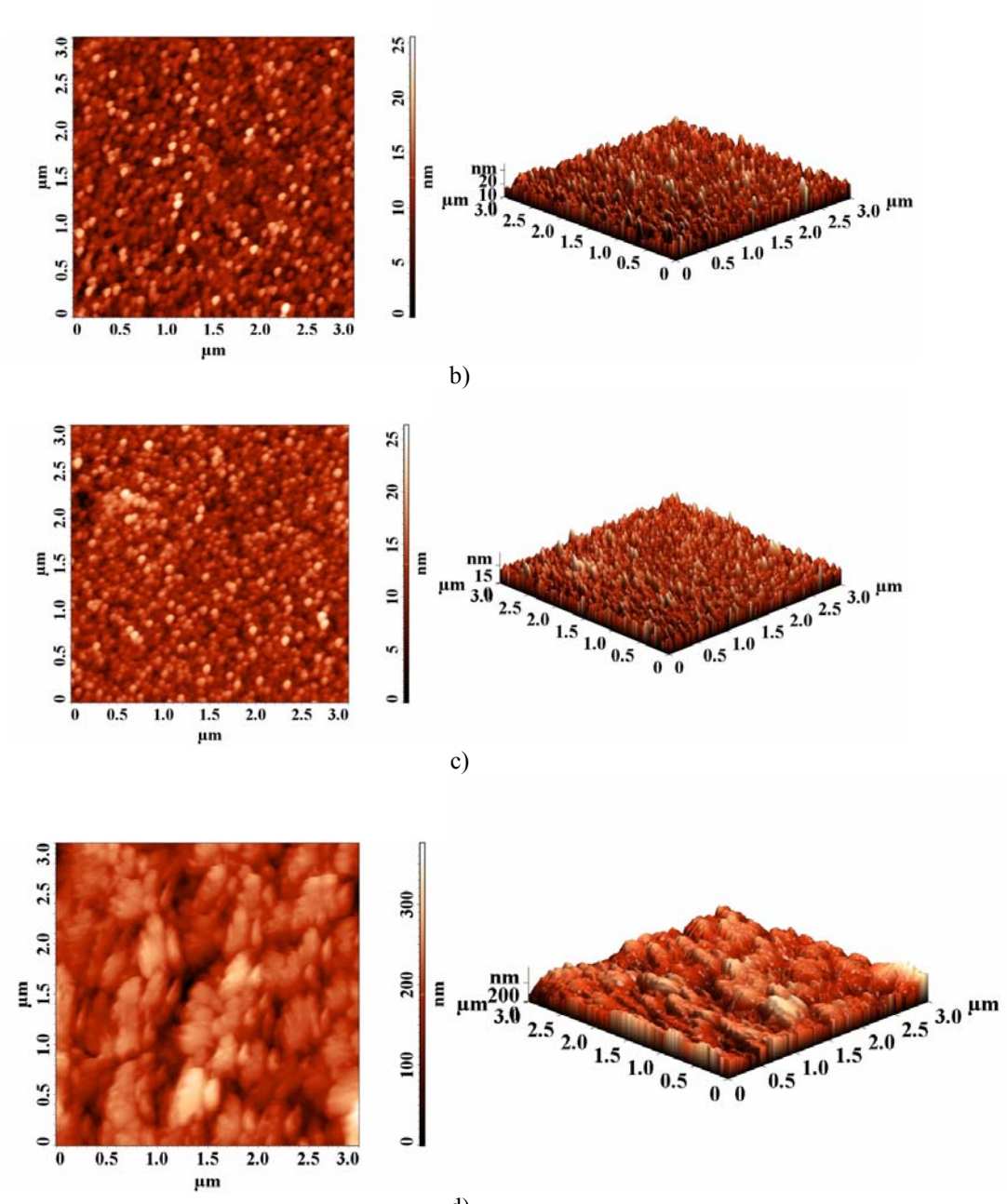


Fig. 3. *2D (left) and 3D (right) images of SnO_2 thin films with thickness of 500 nm obtained at temperatures of 300K (a) 373K (b) 443K (c) and 583K (d)*

Table 2 *Some roughness parameters of SnO_2 thin films*

Substrate temperature	Film thickness	Average roughness, Sa	Root mean square, Sq
[K]		[nm]	
300	500	3.1966	4.06847
373	500	2.31532	2.99308
443	500	2.20535	2.81901
583	500	34.3865	43.9978

Studying the surface topography of SnO_2 thin films thus obtained was found that surface morphology of the substrate is influenced by temperature. Very small values of roughness of SnO_2 nanostructures thin films at temperatures of 300, 373, 443K, and a much increased value at 583K. This different behavior may be due to the amorphous structure of the sample at this temperature, confirmed by the TEM analysis [20, 21].

Because SnO_2 nanostructured thin films have a low electrical conductivity, their research by scanning electron microscopy is difficult. For this reason films were covered with a gold layer with a thickness of 5Å using Edwards AUTO 500 equipment. In this case, the SEM study was performed with an accelerating high voltage of electrons of 10kV and a magnification of 30,000x.

The images presented in fig. 4 show that for the same film thickness of 500nm, deposited layer structure is influenced by the substrate temperature. Thus, Fig. 4a shows the image of a film deposited at 300 K consisting of islands composed of nanoparticles whose size varies between 133.7 to 188.8 nm. At a substrate temperature of 373K, image shown in fig.4b, the coating consists of islands joined together and that includes nanoparticles whose dimensions range from 104.2 to 178.3nm. At a substrate temperature of 443K, Fig. 4c, the thin film tends to appear uniformly encompassing irregular particles of size 133.2-570.3nm arranged disorderly. Films obtained at a support temperature of 583K, fig.4c, are smooth including nanoparticles uniformly arranged with irregular polygonal shapes and sizes of 276.4-660.4nm.

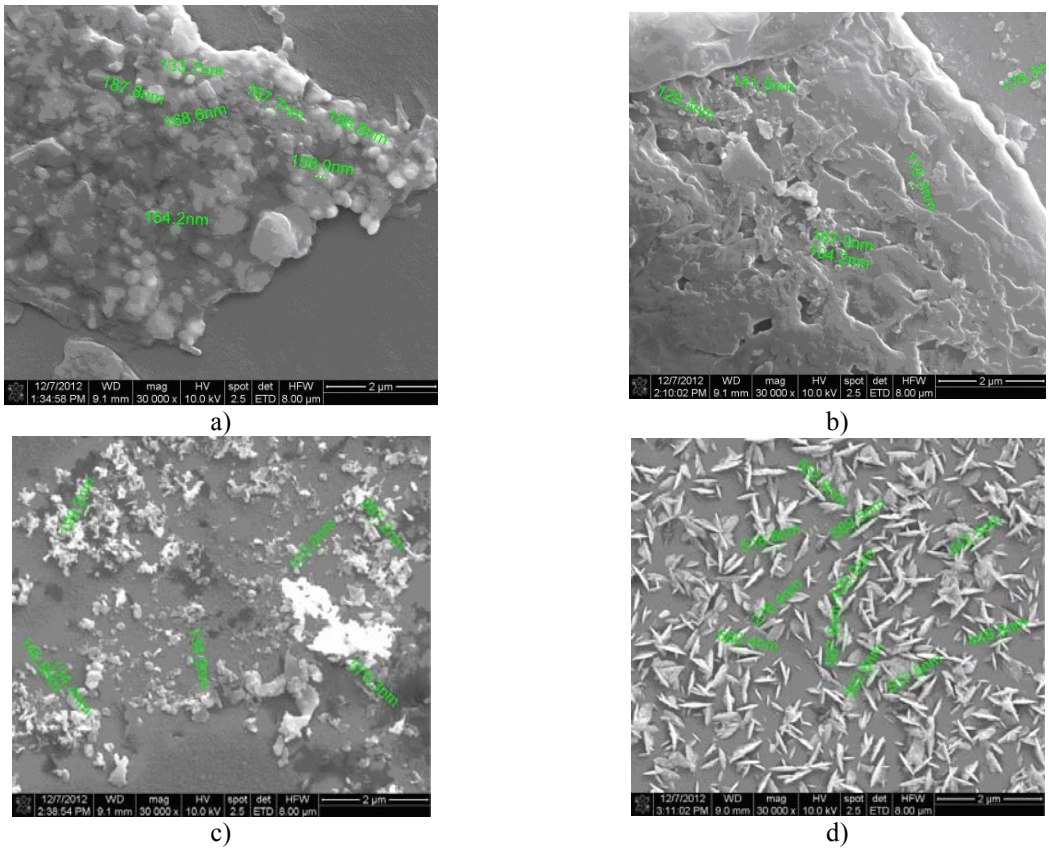


Fig. 4. *SEM images of SnO_2 nanostructured thin films with thickness of 500 nm obtained at temperatures of 300 K (a) 373 K (b) 443 K(c) and 583 K(d)*

4. Conclusions

By EB-PVD method, we prepared SnO_2 nanostructured thin films with thickness 500 nm on glass substrates, at different temperatures of 300, 373, 443 and 583K. We investigated structural and morphological properties of the thin films thus obtained using TEM, AFM and SEM techniques. The sample obtained at 373K has the highest average size of nanoparticles, 40.78nm, while those obtained at 300K have the lowest average size of nanoparticles, 9.48nm. Nanoparticles have a spherical shape and it is not influenced by the change of the substrate temperature. Roughness parameters show different values to changes in the substrate temperature. The greatest roughness parameters of the SnO_2 thin film corresponding to a support temperature of 583K when the film is in amorphous state.

Structural and morphological properties were studied, proving to be very sensitive to the change of substrate temperature during nanostructuring of SnO_2 thin depositions. The substrate temperature strongly influences the structural and the morphological properties during nanostructuring of SnO_2 thin films, which is in good agreement with literature dates [16-21].

Acknowledgements

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