

THE ANNALS OF "DUNAREA DE JOS" UNIVERSITY OF GALATI. FASCICLE IX. METALLURGY AND MATERIALS SCIENCE $N^0.\ 2-2013, ISSN\ 1453-083X$

PREPARATION AND CHARACTERIZATION OF Tin OXIDE THIN FILMS

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ABSTRACT

Tin oxide (SnO) thin films were prepared onto glass substrates by thermal evaporation under vacuum. The substrate temperature was kept constant at 300 K during the film growth. The structural studies using transmission electron microscopy (TEM) analysis showed that the SnO thin films have a polycrystalline and tetragonal crystal structure with preferential orientation of (110) planes parallel to the substrate. Optical transmission and reflection spectra, at normal incidence, in the spectral range 300-1100 nm, are investigated. The optical properties of SnO thin films were determined. The optical energy band gap, $E_{\rm g}$, has been estimated from the absorption coefficient values using Tauc's procedure. It is found that the SnO thin films exhibit direct band gap.

KEYWORDS: thermal evaporation, tin oxide, TEM, optical properties

1. Introduction

In recent years, transparent conducting oxide films have attracted the researchers due to their potential use in a variety of applications including optoelectronic devices, such as solar cells, flat panel displays, light emitting devices and gas sensors [1-5]. Among the transparent conducting oxide films, tin oxide exhibitts interesting physical properties suitable for these applications.

There are different methods of depositing tin oxide thin films, in particular spray pyrolysis [6,7], magnetron sputtering [8], pulsed laser ablation [9], chemical routs by using sol-gel [10] and thermal evaporation technique [11]. In the present work, the ojective was to prepare SnO thin films by vacuum thermal evaporation technique and to study the structural and optical properties of these films.

2. Experimental research

SnO thin films were deposited on highly clean glass substrates by thermal evaporation under vacuum ($p=10^{-5}$ torr) of SnO₂ powder (Sigma

Aldrich, purity 99.99%). The glass substrate was kept at a temperature of 300K during deposition of thin films and the thickness of SnO thin films was measured by interferometric method [12] using a Linnik microscope.

The thickness of investigated samples was d=0.10 μ m. The microstructural characteristics of the SnO thin film such as morphology, particle size and crystal structure were analyzed by transmission electron microscopy (TEM). The TEM investigation was performed on a Philips CM-120 electronic microscope with an accelerating voltage of 120kV.

Optical transmission and reflection spectra were recorded in the wavelength range of 300-1100nm using Perkin Elmer Lambda 35 UV-Vis Spectrometer and UV Win Lab software.

3. Results and discussion

The investigated SnO thin films were adherent, uniform and covered well over the substrate. The electron diffraction was used for determining the structural phases of SnO thin films. Selected area electron diffraction (SAED) patterns were taken from



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selected areas in the SnO thin films. Figure 1 shows the SAED patterns of the SnO thin film deposited at substrate temperature of 373K. Six intense diffraction rings shown are corresponding to diffraction from (101), (200), (220), (211), (321) and (312) planes. The SnO thin film deposited at 300K exhibits the polycrystalline structure and corresponds to a tetragonal rutile structure. The crystallites are preferentially oriented with the (101) planes parallel to the substrate surface. Lattice spacing, d_{hkl} , was calculated using Bragg's equation together with the Miller indices and the calculated values of d_{hkl} are inserted in SAED pattern.

The lattice parameters, a and c, of SnO polycrystalline thin films, with a tetragonal structure, were calculated using the relation [11, 12]:

$$d_{hk\ell} = \frac{1}{\sqrt{\frac{h^2 + k^2}{a^2} + \frac{\ell^2}{c^2}}}$$
 (1)

where h, k and ℓ are Miller's indices corresponding to (hk ℓ) lattice plane. The values of lattice parameters were found to be a=4.75Å and c=3.08Å, respectively. These values are in good agreement with those reported recently in Ref. [6,11,14,15] for SnO thin film.

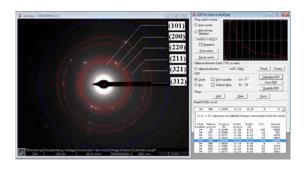
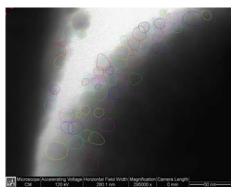


Fig. 1. SAED pattern of SnO thin film

Figure 2(a) shows bright-field transmission electron microscopy (BF-TEM) images recorded from plan view specimens, showing that SnO thin films exhibit a grainy structure which consists of many small grains of relatively uniform size forming morphologically homogeneous films. The size distributions of the crystallites, obtained from measurements on around 400 grains, are plotted in Fig. 2(b).

The distributions of grain sizes, as measured from BF-TEM images, were fitted to the lognormal curves [16,17]. The mean grain size, x_c , was found to be around 12.26nm (Fig. 2b).



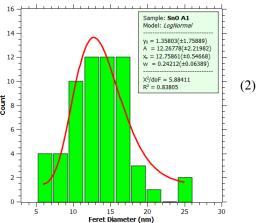


Fig. 2. (a) BF-TEM images of SnO thin films, (b) Grain size distribution of the SnO thin film

Figure 3 shows the optical transmittance (T%) and reflectance (R%) spectra of SnO thin films deposited at the investigated substrate temperature. The thin films exhibit more than 75% of transmittance in the visible and near-infrared wavelength regions with some interference fringes and sharp ultraviolet absorption edges. Also, in Fig. 3 it can be seen that all samples show reflectance more than 25% the for whole investigated wavelength range.

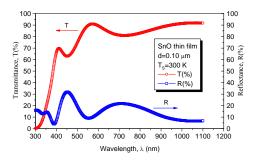


Fig. 3. Transmission and reflection spectra of SnO thin film



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The absorption coefficient, α , was calculated by relation [18-20]:

$$\alpha = \frac{1}{d} \ln \frac{(1 - R^2)}{T} \tag{2}$$

where d=0.10 μm is the film thickness, T the transmittance and R reflectance of the SnO thin film. The variation of absorption coefficient with wavelength is shown in Fig. 4.

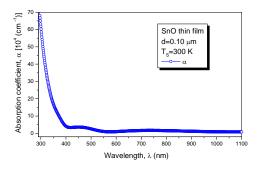


Fig. 4. Variation of absorption coefficient of SnO thin film

The optical band gap energy, E_g , can be estimated from the fundamental absorption edge of SnO thin films. Assuming the allowed direct transition between valence and conduction band, the values of optical band gap of SnO thin films were determined from the dependence of absorption coefficient, α , on the photon energy, hv, using Tauc's relation [20-22]:

$$\alpha h v = A \left(h v - E_g \right)^{1/2} \tag{3}$$

where A is a parameter that depends on the transition probability. Figure 5 exhibits the plots of $(\alpha h \nu)^2$ versus photon energy, hv. The E_g values are determined by extrapolating the linear portion of the curves to the hv axis. The obtained value of optical band gap energy was found to be 3.78eV; which is in good agreement with those found in literature for this kind of thin films [8,10,23-28].

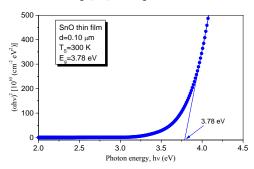


Fig. 5. The $(\alpha h v)^2$ versus (h v) plot for investigated SnO thin film

4. Conclusions

SnO thin films were deposited at substrate temperature of 300 K using the thermal evaporation technique. The TEM patterns showed that the films is polycrystalline with tetragonal rutile structure. The obtained values of lattice parameters were: a=4.75Å and c=3.08Å, respectively. The mean grain size was found to be around 12.26nm. Transmission and reflection spectra in the wavelength range 300-1100 nm were used to calculate the absorption coefficient and optical band gap energy evaluated from absorption spectra was found to be 3.78 eV which is in good agreement with literature.

References

[1]. A.L. Dawar, J.C. Joshi - J. Mat. Sci. 19, 1, (1984).

[2]. B. Russo, G.Z. Cao - Appl. Phys. A. 90, 311, (2008).

[3]. P.S. Patil, S.B. Sadale, S.H. Mujawar, P.S. Shinde, P.S. Chigare - *Appl. Surf. Sci.* 253, 8560, (2007).

[4]. C.M. Lampert - Solid Energy Mater. 6, 1, (1981).

[5]. D.S. Lee, Y.T. Kim, J.S. Huh, D.D. Lee - *Thin Solid Films* 416, 271, (2002).

[6]. P.S. Shewale, S.I. Patil, M.D. Uplane - Semicond. Sci. Technol. 25, 115008, (2010).

[7]. E. Elangovan, S.A. Shivashankar, K. Ramamurthi - J. Cryst. Growth 276, 215, (2005).

[8]. D. Beena, K.J. Lethy, R. Vinodkumar, V.P. Mahadevan - Sol. Energ. Mat. C. 91, 1438, (2007).

[9]. J. Montero, J. Herrero, C. Guillen - Sol. Energ. Mat. C. 94, 612, (2010).

[10]. B. Benrabah, A. Bouaza, S. Hamzaoni, A. Dehbi - Eur. Phys. J. Appl. Phys. 48, 30301, (2009). (4)

[11]. A. Sharma, D. Prakah, K.D. Verma - Optoelecton. Adv. Mat. 1(12), 683, (2007).

[12]. C. Gheorghies, L. Gheorghies, R. Chirila Roentgenocristalografie, Ed. Tehnica, Bucuresti, (1994).

[13]. K. L. Chopra - Thin Film Phenomena, McGraw-Hill, New York Book Co. 101, (1969).

[14]. S. Sambhaji, S. Bhande, Gauri A. Taur, Arif V. Shaikh, Oh-Shim Joo, Myung-Mo Sung, Rajaram S. Mane, Anil V. Ghule, Sung-Hwan Han - *Mater. Lett.* 79,29, (2012).

[15]. Y. Liu, Y. Dong, G. Wang - Appl. Phys. Lett. 82(2), 260, (2003)

[16]. N. Tigau, V. Ciupina, G. Prodan, G.I. Rusu, E. Vasile - J. Cryst. Growth 269, 392, (2004).

[17]. M. Huang, Y. Wang, Y. Austin Chang, *Thin Solid Films* 449, 113, (2004.

[18]. F. Aousgi, M. Kanzari - Energy Procedia 10, 313, (2011).

[19]. G.I. Rusu, M.E. Popa, G.G. Rusu, Iulia N. Salahoru - Appl. Surf. Sci. 218, 213, (2003).

[20]. T. S. Moss, M. Balkanski, (eds) - Handbook on Semiconductors, Optical Properties of Semiconductors, North-Holland, Elsevier, Amsterdam, (1994).

[21]. N.F. Mott, E.A. Davis - Electronic Processes in Non-Crystalline Materials, Clarendon Press, Oxford, (1971.

[22]. N. Tigau, V. Ciupina, G. Prodan - J. Cryst. Growth 277, 529, (2005).

[23]. S. Condurache-Bota, N. Tigau, A.P. Rambu, G.G. Rusu, G.I. Rusu - *Appl. Surf. Sci.* 257, 10545, (2011).



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[24]. H. R. Fallah, M. Ghasemi, A. Hassanzadeh, H. Steki -Physica B 373, 274, (2006).
 [25]. S. S. Lekshmy, L.V. Maneeshya, P.V. Thomas, K. Joy -

Indian J. Phys. 87, 33, (2013).

[26]. N. F. Habubi, K.A. Mishjil, S.S. Chiad - *Indian J. Phys.* 87(3), 235, (2013).

[27]. C.Gheorghies, L. Gheorghies - Nanomateriale si Nanotehnologii, Ed. CERMI, Iasi, (2008)

[28]. L. Gheorghies, A. Sion - Obținerea și analiza acoperirilor metalice de protecție, Ed. CERMI, Iași, (2012).