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Synthesis and characterization of nanocrystalline SnO_2 film by sol-gel assisted screen-printing route

ABSTRACT

Generally, the sol–gel route engages spin-coating or dip-coating for the film deposition on a substrate. This paper first time establishes the synthesis of SnO_2 film using an innovative and economical sol-gel assisted screen-printing route. As deposited film was finally sintered at 450 °C for 10 min. The polycrystalline nature and single-phase tetragonal structure of SnO_2 was confirmed by XRD result. SEM result exposes the homogeneous pattern of different size particles with porous nature. Diffuse reflectance spectrum (DRS) analysis shows the band gap of sintered SnO_2 film to be 3.65 eV. The Hall measurement test conveys the n-type conductivity for the film having a resistivity of 1.07 x 10^{-3} Ω cm.

Keywords:Sol-gel, film, XRD, Diffuse reflectance*

1. INTRODUCTION

Materials in thin films form serve as the main component of photovoltaic technology, functional nano devices and advanced electronics [1].Transparent conductive oxide (TCO) films (ZnO, SnO_2, and TiO_2) earned significant recognition due to their applicability in optical and optoelectronic devices [2]. These films exhibit less electrical resistivity and greater optical transmission [3]. Among these TCO's, tin oxide (SnO_2) is an important n-type semiconductor that has a wide band gap (3.6 eV) and large electron mobility (200 $cm^2 V^{-1} S^{-1}$) [4]. In thin film form, SnO_2 was used in diverse applications like solar cells, optoelectronic devices, photo-catalytic devices, and gas sensors [4]. Numerous deposition routeswere described to obtain SnO_2 thin films, such as sol-gel spin-coating [1], sol-gel dip- coating method [2], atomic layer deposition [3],Co-precipitation method [4], Spray pyrolysis method [5], pulsed laser deposition [6].

This paper demonstrated for the first time the synthesis of SnO_2 film via an innovative sol-gel assisted screen-printing route. This is an ecological as well as low-cost route and appropriate for coating huge area [7, 8].Uniform mixing of composition at the atomic level can also be achieved via this route [7].Therefore, this investigation is carried out with keen enthusiasm to study the synthesis and properties of sol-gel assisted screen-printed synthesized SnO_2 film.

2. EXPERIMENTAL

Using a sol-gel assisted screen-printing route, SnO_2 film was successfully synthesized on clean glass substrate. The starting materials taken were tin Chloride dehydrate ($SnCl_2$. $2H_2O$: as source material), absolute ethanol (as binder) and monoethanolamine (as stabilizer). Tin chloride dehydrate was dissolved in 20 ml of absolute ethanol, then a few drops of monoethanolamine were added to stirred solution for stabilization. The obtained solution was vigorously stirred at 70 ^{0}C for 2 hours. Finally, the resulting gel of SnO_2 was left under the room temperature for 48 hours. The gelated sol (sol) was coated on glass substrate

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using a screen-printing setup whose pictorial representation is depicted in fig.1. The coated glass substrate was heated at 100 ^{0}C for 10 min to burn the volatile solvents from the film. Further to get rid of unwanted organic residues, the film was fired (sintered) at an optimized temperature of 450 ^{0}C for 10 min.

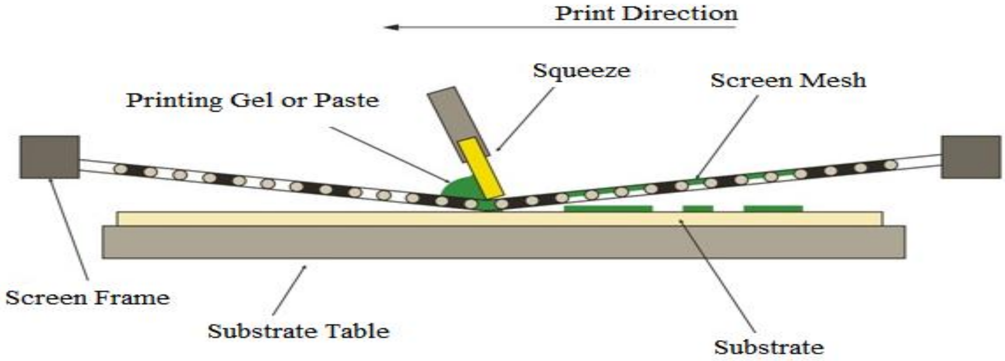

Figure 1. Pictorial representation of screen-printing setup

The thickness of screen-printed film of SnO_2 was measured using mass difference method. The thickness of SnO_2 film was observed to be 6 µm. The structural study of the film was made using XRD (Bruker D8-Advance) measurement. The microstructure of the SnO_2 film was scrutinized via a scanning electron microscope. The UV-Vis diffuse reflectance spectrum (DRS) for the film was obtained on a spectrophotometer (Hitachi U-3900). Electrical transport study was carried out on Hall measurement set up.

3. RESULTS AND DISCUSSION

3.1. Structural study by XRD

XRD profile of sintered SnO_2 film is publicized in fig.2. The XRD profile shows intense peaks suggesting the polycrystalline nature of the film. The peaks indicate that the SnO_2 film crystallizes in tetragonal structure (JCPDS-41-1445) and matches well with the literature [9,10].

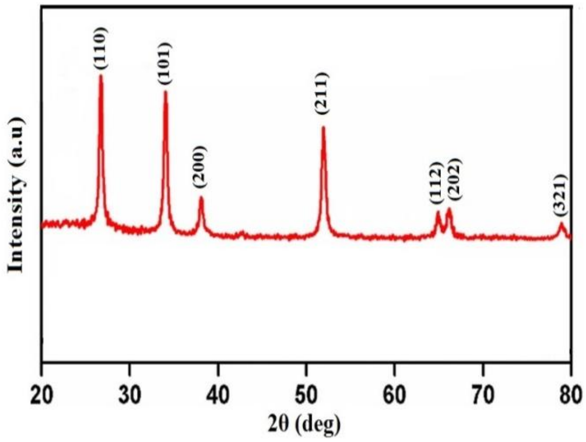

Figure 2.XRD pattern for sol-gel aided screen-printed SnO_2 film

The film shows the peaks related to (110), (101), (200), (211), (112), (202) and (321) planes respectively. The film favours the growth along (110) plane. The average particle size (D) for SnO_2 film estimated by the classical Scherrer-Debye formula was 60 nm.

3.2. Morphology study by SEM

SEM study was done for analysis of surface morphology of the SnO_2 film. Fig. 3 shows the scanning electron microscope (SEM) micrograph of sintered SnO_2 film. It was inferred from SEM micrograph that the particles of different size were arranged in homogeneous pattern on the surface with porous structure. Almost similar surface morphology was reported earlier for SnO_2 film by Miskovic et al. [11].

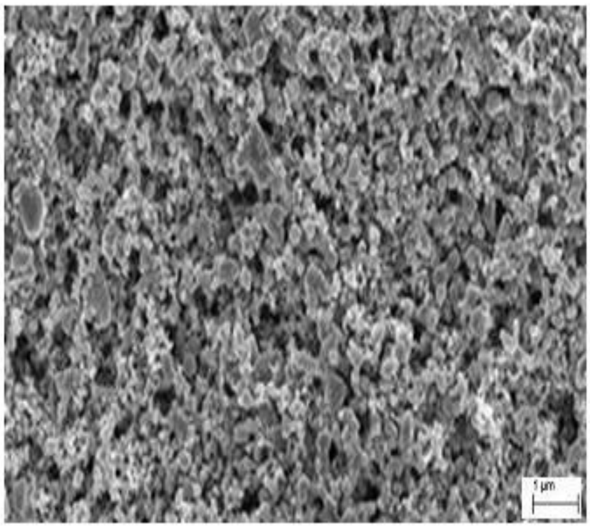

Figure 3.SEM image for sol-gel aided screen-printed SnO_2 film

3.3.Optical study by UV-VIS Diffuse reflectance spectroscopy

UV-Vis study is a productive tool to find out the band gap of a material. To find out the band gap of sol-gel assisted screen-printed synthesized SnO_2 film on a glass substrate, the reflection spectra in wavelength range of 200-700 nm was measured and is presented in fig.4. This spectra depicts a strong decrease after 340 nm, which is due to optical transition taking place in band gap. To precisely find the value of band gap, the Kubelka-Munk (K-M) approach [Equation (1)][10,12] was applied:

$$F(R) = \frac{(1-R)^2}{2R} \quad (1)$$

whereF(R): is Kubelka-Munk function and R: is reflectance.

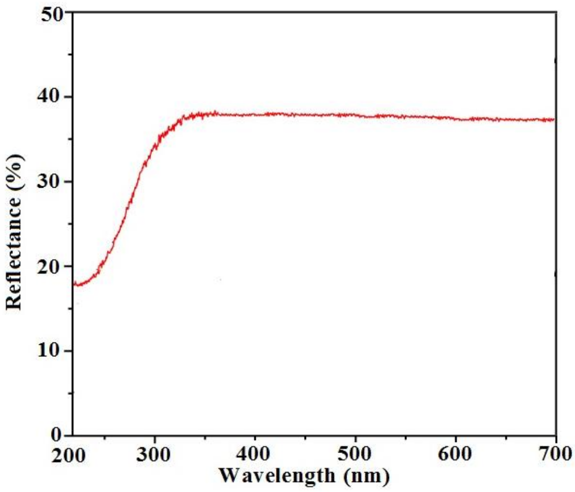

Figure 4.Diffuse reflectance spectra for sol-gel aided screen-printed SnO_2film

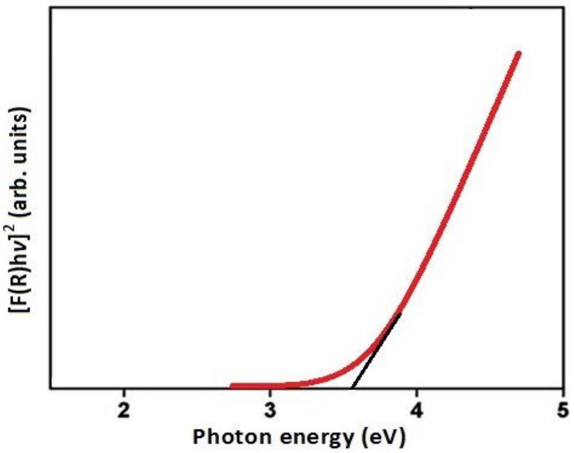

Figure 5.Plot of $[F(R)hv]^2$ versus (hv) for sol-gel aided screen-printed SnO_2 film

To find the band gap from this approach (K-M approach), $[F(R)hv]^2$ on y-axis versus (*hv)* on x-axis variation curve was plotted as given in Fig. 5. Band gap value of 3.65 eV was found from the extrapolation of the straight (linear) part of this curve on x-axis [13]. The band gap (3.65 eV) of the synthesizedSnO_2 film is slightly larger than the band gap of bulk SnO_2 (3.6 eV) may bedue to effect of quantum confinement [14].The found band gap energy is in excellent agreement with the reported one found by other researcher group for SnO2 film synthesized with different method [15].

3.4.Electrical study by Hall measurement

Hall measurement test for synthesized SnO_2 film was done at room temperature and the test shows that film has n-type conductivity. The n-type conductivity of SnO_2 film was ascribed to the emergence of shallow donor levels taking place due to existence of oxygen vacancies and interstitial tin in the SnO_2 lattice [1].The mobility, resistivity and carrier concentration obtained from Hall measurement data for the synthesized SnO_2 film was 67 cm^2/ Vs, 1.07 x $10^{-3}\Omega$ cm and 8.4 x 10^{20} / cm^3 respectively. A similar order of resistivity for SnO_2 film was recently reported by Kiruthiga et.al.[16].

4. CONCLUSIONS

SnO_2 film was successfully accomplished through sol-gel assisted screen-printing processing. XRD study reveals the creation of poly-crystalline, single phase and tetragonal structure of SnO_2. SEM study indicates the particles of different sizes are homogeneously distributed on the complete surface of the film. The band gap of SnO_2 film was found to 3.65 eV. n-type conductivity and a resistivity of 1.07 x 10^{-3} Ω cm were revealed for the film via the Hall measurement test. The extracted results go a long distance to show that goodquality SnO_2 films can be synthesized via a simple, innovative, low-cost and environment friendly sol-gel assisted screen-printing processing.

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IZVOD

SINTEZA I KARAKTERIZACIJA NANOKRISTALNOG SnO_2 FILMA PUTEM SITOŠTAMPE UZ POMOĆ SOL-GELA

Generalno, sol-gel postupak uključuje centrifugiranje ili potapanje za taloženje filma na podlogu. Ovaj rad po prvi put uspostavlja sintezu SnO_2 filma koristeći inovativnu i ekonomičnu rutu sitoštampe uz pomoć sol-gela. Pošto je taložen, film je konačno sinterovan na 450°C tokom 10min. Polikristalna priroda i jednofazna tetragonalna struktura SnO_2 potvrđena je XRD postupkom. SEM rezultat otkriva homogeni obrazac čestica različite veličine, porozne prirode. Analiza spektra difuzne refleksije (DRS) pokazuje da je velicina u pojasu sinterovanog SnO_2 filma 3,65eV. Holov test merenja prenosi provodljivost n-tipa za film koji ima otpornost od 1,07 k 10^{-3}Ωcm.

***Ključne reči**: sol-gel, film, XRD, difuzna refleksija*

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