IRON DOPING SnO₂ THIN FILMS PREPARED BY SPRAY WITH MOVING NOZZLE: STRUCTURAL, MORPHOLOGICAL, OPTICAL, AND TYPE CONDUCTIVITIES

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Elaboration of 0-10 wt.% Fe-doped SnO₂ thin films using spray pyrolysis technique on 480°C heated glass with moving nozzle. Tin(II) chloride SnCl₂ and iron(III) chloride FeCl₃ were employed as sources of SnO₂ and Fe doping, respectively. The structural, opticaland electrical characteristics of pure SnO₂ and Fe-doped SnO₂thin films with different ratios weight Fe/Sn were studied. Transmittance spectra of all the thin films display high transparency more than 78% in the visible region. The optical gap of the thin films was found to be in 3.56-3.80eV range. In general, XRD shows polycrystalline structures with cassiterite tetragonal crystal phase with apreferential orientation for non-doped SnO₂ toward (*211*) plane then shifts from (*211*) to (*110*) planes for 1, 2 and 4 wt. %. The values of crystallite size, along the preferred orientation, were found to be in 36.11-56.99nm range. SEM survey of the surface morphology shows that the whole substrates are well covered by SnO₂ uniform surface with few numbers of pores. Those pores density increases with increasing iron doping.Electrical resistivity increases from 1.2×10^{-4} to $3.4 \times 10^{-3}\Omega$ cm.The Hall effect measurements have shown n-type for pure SnO₂ then the type conductivity changes to p-type for Fe-doped SnO₂ thin filmsover 2 wt.%.

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1. Introduction

Tin dioxide (SnO_2) exists in nature as the mineral cassiterite under its rutile unit cell with tetragonal crystal structure[1]. As known, SnO_2 isann-type semiconductor, which has a broad band-gap ($E_g>3.6eV$) and high conductivity. Like other binary metal oxides such as ZnO, In_2O_3 , oxygen vacancies are responsible to generate electron-carrier for conduction in SnO_2 [2-4]. SnO_2 thin films utilized innumerous direct applications namely as semiconducting gas sensors[5], solar cell [6], photo-catalysis [7], light emitting diodes (LEDs), electrochromic displays (ECDs), and many more[8]. Several elaboration methods have been developed to get tin oxide thin films deposition.

Physical and chemical are the most two trends to elaboratethin films products. For physical one thereare various methods employed for deposit SnO_2 thinfilm: physical vapordeposition (PVD)[9], pulsed laser ablation[10], molecular beam epitaxial [11] and so on. Whereas for the chemical trend, there is also many methods for instance spray pyrolysis[12], solgel, dip/spin coating method[13]which employ precursor solutions. Spray pyrolysis is a technique having great consideration to prepare thin and thick films, and powders. This method is simple and relatively cost-effective processing[14]. Unfortunately, this method spends more time in elaborating samples because it needs a stay in time to elevate the substrate reaction temperature, so

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it is necessary to give itamelioration moving the spray nozzle (*i.e*to preserve the substrate temperature constant).

Elements doping tin oxide can significantly improve its properties, for example, adding fluorine (F)[10, 15,16] and Antimony (Sb)[17-19]can ameliorate its factor of merit.For Fe-doped SnO₂ thin films, there are few works have been done. M.-M. Bagheri-Mohagheghi et al [20]studied thermoelectric properties.M. C. Rao et al [21]found that iron take place at a distorted octahedral site symmetry into the host lattice.B. Majibul Haque et al [22]interested the influence of Fe^{+2}/Fe^{+3} ions in tuning the optical gap of tin oxide thin films without taking in count the electrical properties. In this context Fe-doped tin oxide topic needs to be further investigated.

This work aims to study the effect of iron (Fe) doping concentration (0-10wt.% Fe/Sn) on the structural, optical and electrical properties of SnO2 thin films in the precursor using spray pyrolysis with moving nozzle[23]. For this X-ray diffraction (XRD), UV-Vis absorption and FT-IR spectroscopy, scanning electron microscopy (SEM), four-point probe and Hall effect were used for physical properties measurements.

2. Experimental methods

2.1. Solutions and thin films preparation

0.5M solution of tin dichloride (SnCl₂, 2H₂O) were used for preparing SnO₂thin films. The precursor was liquefied in bi-distilled water and methanol in 1:1 volume ratio with adding few drops of (HCl). The starting solutionwas obtained by dissolving mass weight of SnCl₂ (m =2.2564g) in volume (V = 20ml ofbi-distilled water and methanol). Iron doping was achieved by adding anappropriate amount of iron chloride (Fe Cl₃) dissolved in bi-distilled water to the mother solution in respect to 0-10 wt.% iron concentrations in mixture solution. The mixture was agitated at room temperature for 30 minutes to yield a clear and transparent solution. The gained mixture was utilized as a stock solution for spray.Glass slides of microscopic (Ref 217102) were employed as substrates. Cleaning substrates wereproceed with alcohol in an ultrasonic bath and distilled water then blowing-dried with air. The substrates temperature was fixed at 480°C. The blend solutions were sprayed for 0-10 wt. % Fe doped SnO₂ separately on the heated glass substrates by spray system withamovingnozzleto avoid the fall down of temperature on the surface substrate.

2.2. Characterization

The structural characterizations are accomplished using an X-ray diffractometer (BRUKER - AXS type D8) armed with X'Pert High Score working underCu K α ($\lambda = 1.5405$ Å) radiation and a range of (2θ =20-80°). The crystalline size of the thin films was valued using X'Pert High Score. Morphological properties were achieved with the help ofscanning electron microscope (SEM) type FEIQuanta 250.Optical transmission spectra were obtained using an UV-visible spectrophotometer (Shimadzu, Model 1800) in spectral region 200-900nm. The other different optical constants such as Urbach energy and the optical band gap were estimated using data and fitting of the transmission spectrum; whereas the film thickness (*t*) was carried out using simulation program. The chemical bonds were found by using FT-IR spectroscopy (Shimadzu IR-Infinity1).The electrical properties, mainly, the resistivity of thin films was characterized using four points probe method on 1 × 1 cm² sample sheets. The nature of majority carrier concentration was deduced from the Hall voltage.At room temperature (*rt*), measurements were done.

3. Results and discussion

3.1. Structural properties

X-ray diffractionspectra of SnO₂ thin films, elaborated by spray pyrolysis with different Fe/Sn concentration in the precursor, are shown in Fig.1.As can be seen, the diffraction peaks were observed at $2\theta = 26.67^{\circ}$, 33.344° , 38.035° , 51.816° , 54.712° , 61.886° , 65.898° , and 78.597° which are related to the following planes (*110*), (*101*), (*200*), (*211*), (*220*), (*310*), (*301*), and (*321*), respectively. The obtained XRD spectra accorded well with the space group P42mnm according to

tetragonal rutile SnO₂ structure having JCPDS card: (No.041-1445)[24] and showed the polycrystalline nature of the thin films. The non-doped and Fe doped SnO₂ thin films grow along (211) as preferred orientation. There was a decrease in peak intensity (211) with an increase in Fe doping until4wt. % Fewhere preferential orientation growth of Fe-doped SnO₂ shifts toward (110) planes. Beyond this doping level, the intensity increases for all the rest amount of doping. The intensities of (211) increase and some others peaks are related to the films enhancement crystallinity. Another weird and wonderful peak which was observed at 2θ = 31.86° indicating the appearance of thecubic system of tin oxide according to JCPDS card: (No. 050-1429)[25]. This peakisrelated to the plan (111) of this system. The peak intensity of (111) evokes its highest value in the case of non-doped film and fall down to reach a minimum at 1wt.% Fe, then starts to increase with Fe doping levels (see Fig. 1).



Fig.1: XRD patterns of 0-10wt.% Fe-doped SnO₂ thin films.

X-ray data were also used for more details concerning the favorite growth. Texture coefficients, TC(hkl), have been evaluated. As well known, TC(hkl) represents the texture of the particular plane, a digression of which from unity implies the preferred growth. TC(hkl) coefficients have been evaluated using the following formula[26]:

$$TC(hkl) = \frac{I(hkl)/I_0(hkl)}{N^{-1}\sum_n^N I(hkl)/I_0(hkl)}$$
(1)

where I(hkl) and $I_0(hkl)$ are the measured intensity and the standard intensity of the same (hkl) plane according to the JCPDS data whereas N and n are the reflection and the diffraction peaks number, respectively. TC(hkl) values of all thin films for (110), (101), (200), (211) and (301) planes upon the doping concentration are exhibited in Fig. 2. As seen the intensities of all peaks were less than unity which confirms the polycrystalline structure of the films.



Fig.2: TC(hkl) variation of 0-10wt.% Fe-doped SnO₂ thin films.

For the tetragonal phase structure, the lattice constants (*a*) and (*c*), were determined from XRD results using the following equation [27]:

$$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2}$$
(2)

Where d_{hkl} ,(hkl),(a) and (c) are the inter-planar space, Miller indices, and the lattice parameters, respectively. The calculated parameters lattice and standard ones are illustrated in Table 1. It was noted that the lattice parameters(a) and (c) of the undoped SnO₂ thin film are (a = 4.721Å and c = 3.181 Å) and decrease after iron doping. Changes in a and c values may be allied to the substitution of Sn⁺⁴ by Fe⁺³. Owing to the difference in their ionic radius (Sn_{i.radius} = 0.71 Å) (Fe_{i.radius} = 0.64Å) [20] The calculated (a) values are slightly fewer than reported in the (JCPDS) card No 41-1445 ($a_0 = 4.737$ Å); whereas, all the values of the lattice constants (c) are strictly less than one ($c_0 = 3.185$ Å) of the same JCPDS card. Moreover, the variations Δa and Δc in the lattices parameters of Fe doped SnO₂ thin films,denoted to the standard ones, are given in Fig.3.



Fig.3: Variations of the lattices parameters, Δa and Δc , for 0-10 wt.% Fe-doped SnO₂ thin films.

Using Scherrer's formula[28], the crystal size of Fe doped SnO_2 thin films, illustrated in Table 1, were calculated from the highly textured peaks:

$$D = \frac{0.9\lambda}{\beta \cos\theta} \tag{3}$$

where D, λ , and β are crystallite size, the X-ray wavelength (1.54056 Å), and the full width at halfmaximum of the most intense diffraction peak; whereas θ is the Bragg angle given in degree.

Fe/Sn	Mean crystalline sizes D (nm)	Lattice constants (Å)				TC(hkl)of plane		
(wt.%)		а	$\Delta a = a - a_0$	С	$\Delta c = c - c_0$	(110)	(101)	(211)
0	50.65	4.721	- 0.0172	3.181	- 0.0061	1.0283	0.7105	1.2547
1	54.50	4.724	- 0.0142	3.181	- 0.0061	1.9243	0.8874	1.7476
2	56.99	4.718	- 0.0202	3.179	- 0.0081	1.5535	1.0655	1.5206
4	36.11	4.723	- 0.0152	3.181	- 0.0061	1.3883	0.7825	1.2432
6	42.46	4.725	- 0.0132	3.181	- 0.0061	1.3821	0.9376	1.482
8	50.46	4.718	- 0.0202	3.179	- 0.0081	0.9266	1.5047	2.1232
10	43.60	4.719	- 0.0192	3.178	- 0.0090	0.6695	1.1005	1.8583

Table1. Crystallite sizes D, lattice parameters (a and c), and texture coefficients TC(hkl) of 0-10 wt.% Fe-doped SnO₂ thin films.

The variation of crystallite size (D) and optical band gap (E_g) as functions of 0-10 wt. % Fe doped SnO₂ are illustrated in Fig.4. It is worth noting that the two amount are inverted.

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Fig. 4.The deferent variation between the crystallite size and optical band gap of 0-10 wt. % Fe-doped SnO₂ thin films.

Fig. 5 displays the features variation of 0-10 wt. % Fe doped SnO_2 thin films crystallite size *D*. Apart the *D* found with (200) which has approximately constant variation, all the *D* variations exhibit the same feature appearance and oscillate around 30nm.



Fig.5.The crystallite size D(nm) along different orientations for 0-10 wt.% Fe doped SnO_2 thin films.

Fig.6shows SEM micrographs of 0, 6, 8, and 10wt. % Fe doped SnO_2 thin films elaborated on a heat glass substrate by spray pyrolysis technique. The surface morphology shows polycrystalline SnO_2 thin films structure with homogeneous surface for the non-doped sample. As well seen a presence of randomly distributed poresfor the doped ones. Those pores increases in density, as the SEM picture illustrated, with doping level. But for the undoped films it is seen that the substrate is well covered by SnO_2 uniform surface with few numbers of pores. SEM images also illustrate a shrink in the size of crystallites after iron doping levels.



Fig.6. SEM pictures of Fe-doped SnO₂ thin films :a) Pure SnO₂, b) 6 wt.% SnO₂:Fe, c) 8wt.%SnO₂:Fe, d) 10 wt.% SnO₂:Fe.

3.2. FT-IRStudies

Fig.7 shows FT-IR spectra of 0-10wt.% Fe doped SnO_2 thin films. In the range 400-1000 cm⁻¹ there is the absorption of (Sn, Fe)-O and Sn-O-Snbands, OH band vibrations in the range 3000-3600 cm⁻¹. The peaks 469 and 768 cm⁻¹ were observed for SnO_2 thin films Fig.7, tow absorption peaks at 471 and 768 cm⁻¹ due to the existence of both the band (Sn-O) and (O-Sn-O) tin oxide functional group[8]. According to the results in further work [29], it can be considered the absorption peaks of 467 and 465 cm⁻¹ for Fe doped tin oxide layers which means there is an overlap between the stretching vibration of Sn-O and Fe-O bonds. The bending vibration of Fe-OH group appears at the 1033 cm⁻¹ peak[30]. This intensity is affected by the adsorbed ament of OH. Based on the X-ray diffraction results of undoped and Fe doped tin oxide layers there are a shift in the diffraction peaks. It indicates a reduction in the size of the unit cell crystalline structure which results from the substitution between Sn^{+4} and Fe^{+3} ions hence Fe^{+3} radius is lower than Sn^{+4} one.



Fig.7. FT-IR transmittance spectra of 0-10wt.% Fe-doped SnO₂ thin films.

3.3. Optical properties

Fig.8 shows UV-visible transmittance spectra of 0-10wt.% Fe-doped SnO_2 thin films. The transmittance of all samples was more than 78% in the entire visible-light region (400-900nm). As

seen in the figure of those spectra, the transmittance features of the films exhibit an increase in their valuesafter doping with iron, reaching 80% at 10 wt.% Fe-doped SnO₂ thin films case.



Fig.8.Transmittance spectra of 0-10wt.% Fe-doped SnO₂ thin films.

Fig. 9exhibits the estimated optical band gap (E_g) of 0-10wt.%Fe doped SnO₂ thin films. E_g values were deduced from transmission spectra using Tauc's relation[31]:

$$(\alpha h\nu)^2 = A(h\nu - E_g) \tag{4}$$

where α , E_g , and hv are the absorption coefficient, the optical band gap of the thin film and the photon energy whereas A is a constant independent on hv. For 0-10wt. % Fe doped SnO₂, E_g values were deduced by extrapolating $(\alpha hv)^2=0$, from the optical transmission and they were ranged in 3.56-3.80eV as recapitulated in Fig.9. The optical band gap values are higher than the value, ($E_g=3.56eV$), reported for single crystal SnO₂[32]. This increase in optical band gap may be due to two effects: the decrease in defects as will be shown in the section of Urbach energy; also it may be related to the increase in the grain size which is inverted to E_g .



Fig.9. Band gap, E_g , estimation from Taucrelation of 0-10wt.% Fe doped SnO₂ thin films.

Urbach energy (E_u) which means the disorder of the structure in the mater of the films is related to the absorption spectrum ccording to[33]:

$$\alpha = \alpha_0 \cdot e^{\frac{h\nu}{E_u}} \tag{5}$$

where α is the absorption coefficient, α_0 is a constant, *h* is Planck's constant,*v* is the radiation incident frequency and E_u is Urbach's energy.

The E_u values are determined by plotting the linear curve $(\ln \alpha)$ variation as function of hv. The inverted slop gives E_u values as shown in Fig.10.Urbach energy values, for 0-10 wt. % Fe doped SnO₂ thin films illustrated in Table 2. It is clear that E_u decreases with increasing rate of doping (0-4 wt. %) and increases until (8 wt. %) then decreases in the rest of the doping level. It is worth seeing that E_u is totally inverted to E_g along the range of doping as exhibited in Fig.11.

Fe/Sn (wt.%)	Thickness (nm)	UrbachenergyE _u (meV)	Band gap E _g (eV)
0	211.29	451.19	3.64
1	204.24	450.01	3.57
2	221.54	325.43	3.80
4	216.94	305.00	3.74
6	215.07	366.19	3.67
8	197.47	604.12	3.56
10	200.51	547.49	3.58

Table 2. Thickness, Urbach energy (E_u) , crystal size, and band gap (E_g) of 0-10 wt. % Fe doped SnO₂ thin films.



Fig. 10. Urbach energy variation for 0-10wt.% Fe doped SnO₂ thin films.



Fig. 11.Band gap (E_g) and Urbach energy (E_u) variations for 0-10wt.% Fe doped SnO₂ thin films.

3.4. Electrical properties

3.4.1. Sheet resistance (R_{sh})

Four probe techniques allow us to carry out the sheet resistance (R_{sh}) values of thin films samples. Fig.12 showsthis resistance R_{sh} variation with the percentage of iron which is obtained from the voltage according to the following formula [34]:

$$R_{sh} = \frac{U}{I} = 4.53 \frac{\rho}{t} \tag{6}$$

where U and I are the applied voltage to the inner probe sample and the outer probe current. ρ and t are the resistivity and thickness of the thin films.

As seen from Fig. 12 that R_{sh} increases from 577.98 Ω/\Box for the undoped to reach15941.51 Ω/\Box for 6wt.% and decrease until 1353.380 Ω/\Box for 8wt.% then return to increase for the last ratio.

C 12000 10000 5000 2000 0 2000 0 2000 0 2000 0 2000 0 2000 0 2000 0 2000 0 2000 0 5 SBO.Fe (wt.%)

Fig. 12. Plot of sheet resistance (R_{sh}) values of 0-10wt.% Fe doped SnO₂ thin films.

3.4.2. Hall effect

Polarity in the Hall voltage (V_{Hall}) is adopted to determine the n or p semiconductors type. When the holes are the majority carrier in p-type semiconductors so V_{Hall} will be positive. Whilst the electrons are the majority in n-type semiconductors then V_{Hall} will be negative. The V_{Hall} and drift velocity (v_x) of holes for a p-type semiconductorcan be written as[35]:

$$V_{Hall} = v_x W B_Z \tag{8}$$

and

$$v_x = \frac{J_x}{ep} = \frac{I_x}{(ep)(Wd)} \tag{9}$$

where e and p, are the amount of the electronic charge and the hole concentration, whereas d and W are the thickness and the width of the sample, respectively. Combining Equations (9) and (8) leads to:

$$V_{Hall} = \frac{I_X B_Z}{ned} \tag{10}$$

The hole concentration is written as:

$$p = \frac{I_x B_z}{e d V_{Hall}} \tag{11}$$

The majority carrier concentration of holes is founded from the magnetic field, current and $\ensuremath{V_{\text{Hall}}}$

For n-type semiconductors, the V_{Hall} and the electrons concentration (n) are according to:

$$V_{Hall} = -\frac{\mathbf{I}_x B_z}{ned} \tag{12}$$

$$n = \frac{I_X B_Z}{e d V_{Hall}} \tag{13}$$

Hall effect measurements of 0-10wt.% Fe doped SnO_2 thin films have been summarized in Table 3.As can be seen there are two branches for which V_{Hall} is positive or negative: the first one

found to be in the range lower than 2wt.% Fe/Sn and the second one is beyond 2wt.%. Based on negative Hall potential found in the case of samples doped lower than 2wt.%, one may proclaim that in this region the samples are n-type whereas for the branch where the potential has positive sign the samples become p-type. The carrier concentrations of the layers are shown in Table 3.

Fe/Sn (wt.%)	$R_{sh}(\Omega/\square)$	Carrier concentration (cm ⁻³)	Type carrier	conductivity (Ωcm) ⁻¹	Resistivity (Ωcm)
0	577.982	-7.00×10^{19}	n	54.00	1.2×10^{-4}
1	1049.592	-8.44×10^{18}	n	28.60	2.1×10^{-4}
2	7943.630	2.38×10^{18}	р	04.35	1.7×10^{-3}
4	7515.560	3.13×10^{19}	р	02.10	1.6×10^{-3}
6	15941.51	$4.60 imes 10^{16}$	р	00.60	3.4×10 ⁻³
8	1353.380	1.60×10^{16}	р	22.80	2.6×10^{-4}
10	2835.500	2.00×10^{18}	р	12.26	5.6×10 ⁻⁴

Table 3.Carrier concentration, mobility, conductivity and resistivity of 0-10 wt. % Fe doped SnO_2 thin films.

4. Conclusions

0-10 wt. % Fe-doped SnO₂ thin films were elaborated by spray with moving nozzle. Several characteristics of 0-10 wt.% Fe/Sn in SnO₂thin films are carried out. Optical transmittance spectra of the thin films are more than78% in visible spectrum. The optical gap of the elaborated thin films were in the range of 3.56-3.80eV. XRD and SEM technics show the presence of polycrystalline structure with cassiterite and tetragonal crystal. The preferential orientation shift from (*211*) toward (*110*) plane for 1,2 and 4 wt.% Fe/Sn only. The values of crystallite size decreases from 56.99to 36.11nm. An increase in electrical resistivity increases from 1.2×10^{-4} to $3.4 \times 10^{-3}\Omega$ cm.

The Hall effect measurements have shown n-type for pure SnO_2 thereafter the type conductivity changes to p-type for more than 2wt. %t Fe-doped SnO_2 thin films. Based on Hall effect, the obtained electrical results of Fe-doped SnO_2 are transparent n-type conducting thin films with iron low doping levels and turns out to be p-type conducting with doping levelsmore than 2wt. % Fe which may be exploited as transparent p-n junction from the same SnO_2 product.

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