# Preparation and characterisation of nanostructured tin oxide (SnO$_2$) films by sol–gel spin coating technique

**E. Savarimuthu[1], N. Sankarasubramanian[2], B. Subramanian[3], M. Jayachandran*[3], C. Sanjeeviraja[4] and S. Ramamurthy[1]**

Highly transparent tin oxide thin films (SnO$_2$) have been prepared using the sol–gel spin coating technique, optimising process parameters such as the solute concentration, spin rate and time of the turn table, film thickness (number of coatings) and heat treatment temperature. The X-ray diffraction (XRD) studies point to the polycrystalline structure of the developed films. The films deposited under optimum conditions are highly transparent in the visible region with a transmittance of 94% at 550 nm and have a resistivity of $3 \times 10^{-2}$ $\Omega$ cm. The films have been obtained at a relatively low process temperature of 400°C.

## Introduction

Transparent conducting tin oxide (TO, SnO$_2$) thin films have been attracting increasing interest as they present many important applications such as liquid crystal displays, opto electronic devices, solar cells, gas sensors, protective coatings, resistors and coating for energy saving windows.[1,2] A number of methods such as chemical vapour deposition,[3–5] sputtering,[6–8] electron beam and reactive evaporation,[9–12] spray pyrolysis[13–16] and sol–gel spin coating[2,17] have been used by several investigators to develop tin oxide films. The film properties are strongly dependent on the crystal structure and the stoichiometry of oxygen and hence dependent on the preparation process and therefore on the process parameters involved.[1,18–21]

Of the various methods, the sol–gel spin coating technique is capable of giving films of good uniformity and better controlled composition and doping level, and does not require any expensive and sophisticated equipment.[2,17,19] So in the present work an attempt has been made to use the spin coating technique through the sol–gel route to prepare tin oxide thin films for device applications using as minimum a concentration of tin precursor as possible.

The spin coating technique using sol–gel form of processing of the coating solution involves several process parameters. The precursor solute concentration, gelation stage of the sol–gel, the turn table spin rate and time and the heat treatment temperature, time and atmosphere are the process parameters to be considered in the spin coating technique.

Investigation of the influence of solute concentration, spin rate, spin time and heat treatment temperature on the film characteristics is essential to obtain definite control of the opto electronic properties of the films. This procedure helps in developing quality conducting oxide films for device applications.[2] In the present work such investigations to study the influence of the spin coating parameters on the quality of the TO films have been carried out and the process parameters are optimised to obtain device quality tin oxide thin films.

## Experimental

Tin oxide thin films have been developed through sol–gel route using an indigenously developed microcontroller based spin coating unit. The solution was prepared by dissolving the desired quantity of stannic chloride (SnCl$_4$.5H$_2$O) in 100 mL ethanol with the addition of some quantity of HCl. This solution is refluxed for a specified time of, say, 1 h. Now the sol is ready. It is kept in an open beaker to gelate and the sol–gel is used for obtaining the coatings. Using the prepared sol–gel, 8 consecutive coatings have been made on 2·5 × 2·5 cm microscope slide glass plates. Each layer has been dried and subjected to heat treatment separately in order to avoid cracking.[16] The volume of HCl in the precursor solution, the refluxing temperature and time have been fixed as 10 mL in 100 mL ethanol, 60°C and 1 h respectively and the other process parameters have been optimised. The prepared SnO$_2$ films have been characterised using JEOL JDX 803a X-ray diffractometer (XRD), Hitachi S 3000 H scanning electron microscope (SEM), Nanoscope E scanning probe microscopy 3138 J for atomic force microscopy

[1]Department of Physics, Gandhigram Rural Institute, Deemed University, Gandhigram 624 302, India
[2]Department of Physics, Thiagarajar College of Engineering, Madurai 625015, India
[3]Central Electrochemical Research Institute, Karaikudi 630 006, India
[4]Department of Physics, Alagappa University, Karaikudi 630 003, India

*Corresponding author, email jayam54@yahoo.com
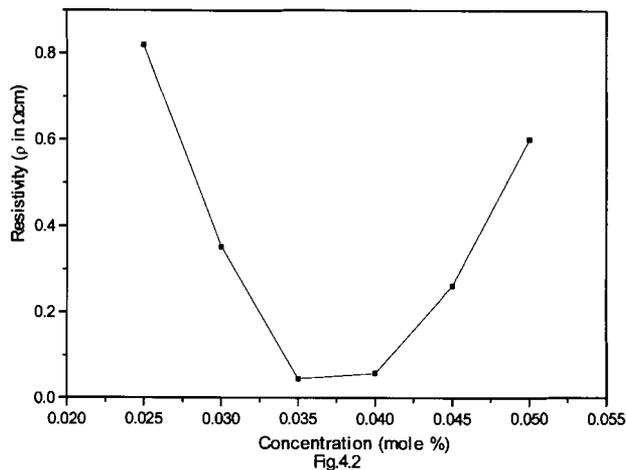
1 **Influence of solute concentration on resistivity of TO films**



2 **Variation of *a* viscosity and *b* density of tin chloride solution with number of days of gelation**

(AFM). The optical transmission and electrical resistivity measurements were carried out using Perkin Elmer Lambda 35 UV-Vis spectrometer and four probe resistivity instrument respectively.

## Results and discussion

### Fixing solute concentration

A concentration of 0·06 mol.% of precursor salt, i.e. stannic chloride $(SnCl_4.5H_2O)$ was used and the sol–gel was prepared. The $SnO_2$ films prepared with a heat treatment temperature of 400°C had powdery layer formation and the films showed a sheet resistance of infinity. This powdery layer could not be removed fully by gentle wiping. When the concentration of stannic chloride was lowered to 0·055, 0·05 and 0·045 mol.%, the powdery layer formation was reduced progressively. At 0·040 mol.% there was no powder formation at all and uniform $SnO_2$ films with good lustre having resistivity values of ~0·05 $\Omega$ cm could be obtained. When the solute concentration was reduced to 0·035 mol.%, uniform good film formation was available and a minimum resistivity of 0·04 $\Omega$ cm was obtained. Further lowering of the solute concentration resulted in films of progressively higher resistivity only, though film uniformity and higher transparency were still retained. The variation of resistivity with precursor solute concentration is shown in Fig. 1.

When the solute concentration increases, the number of atoms (and therefore the crystallite size) and hence the number of oxygen vacancies may increase (for the same number of heat treatment steps) and hence the conductivity rises and therefore the resistivity falls. This will continue until the solute concentration is such that the numbers of atoms are optimum for the film and hence the numbers of oxygen vacancies are optimum at which stage the conductivity will be maximum and the resistivity will be minimum. For the sol–gel spin coated $SnO_2$ films in the present study, this happens at a precursor solute concentration of 0·035 mol.% where a minimum resistivity occurs. At still higher solute concentrations, there appears to be excess solute, which is deposited as a powdery layer on the surface, giving rise to practically zero conductivity, and hence infinite resistivity. This happens beyond 0·040 mol.% solute concentration. So a value of 0·035 mol.% for the
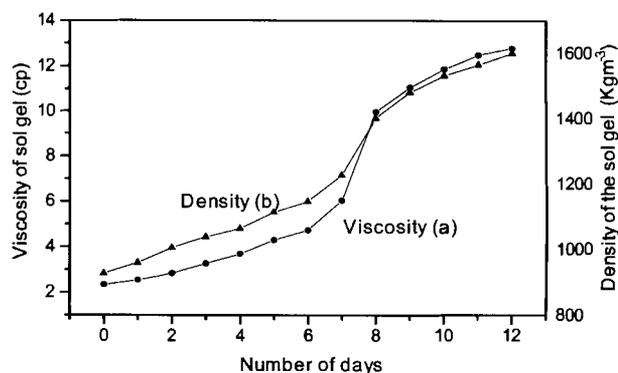
precursor salt stannic chloride (12·27 gm) in 100 mL ethanol has been fixed for the present work.

## Optimisation of gelation time/aging time

The variation of viscosity of precursor sol with gelation time/aging time has been studied and the results obtained are shown in Fig. 2*a*. After the second day of aging, the viscosity of the sol rises, showing that the gelation process has set in, and the rise is uniform up to the sixth day of aging; between 7 and 10 days of aging, the viscosity abruptly rises and after 12 days it tends to a saturation value indicating that gelation stage is complete and thick gel is formed. After 6 days of aging (viscosity of sol–gel >4·5 cp) the coating on the glass substrate becomes less uniform and at higher viscosities streaks of coatings are obtained which progressively becomes more and more irregular; after 12 days of aging the material has been completely thrown off the spinning substrate owing to high viscosity, leading to poor adherence.

Uniform coating could be obtained using the sol–gel of 2–6 days of aging, i.e. with viscosities 2·5–4·5 cp. Below 2 days, the coating is ultra thin. Lin and Wu,[2] from their TEM studies, have established that the particle size grows with gelation time/aging time. So perhaps very heavy particles corresponding to near complete gelation stage are unable to adhere to the substrate and perhaps only when the viscosity is between 2·5–4·5 cp (corresponding to 2–6 days of aging) the particle size is optimum for developing good uniform films.

The variation of density of the sol with aging/gelation time is presented in Fig. 2*b* along with the viscosity variation curve. The figure shows that the density of the sol also follows the same trend as viscosity of the sol with respect to the aging/gelation time and that they are complementary to each other. So either the density or viscosity variation of the sol with aging/gelation time could be used for gelation stage optimisation. Thus 2–6 days of aging time have been taken as optimum for developing tin oxide films. In the present study a gelation time of 3 days has been used for preparing the TO films.

### Selection of turntable spin rate

One of the parameters influencing the quality of thin films in the spin coating method is the spin rate of the turntable. The spin rate affects the degree of centrifugal force applied to the sol, velocity and characteristic turbulence of the air immediately above it. Film
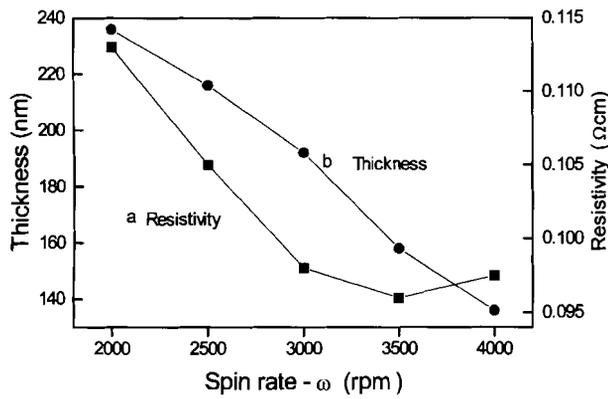
**3 Variation of resistivity and thickness of TO films with turntable spin rate $\omega$**

thickness is largely a balance between the force applied to shear the sol towards the edge of the substrate and drying rate, which affects the viscosity of the sol. As the sol dries, the viscosity increases until the radial centrifugal force of the spin process can no longer appreciably move the sol over the surface. At this point, the film thickness will not decrease significantly with increased spin rate. So the TO films have been prepared with various spin rates varying from 2000 to 4000 rev min$^{-1}$ in steps of 500 rev min$^{-1}$. The variation of the resistivity and the thickness of the TO films with respect to turntable spin rate are shown in Fig. 3.

At relatively low spin rates of 2000 rev min$^{-1}$, formation of yellowish white islands was seen in the films resulting in the poor lustre and the uniformity of the films were rather poor. This may be attributed to the irregular outward flow of the sol, owing to lower degree of centrifugal force acting on the sol, which causes the non-uniform thinning of the sol on the coating surface. When the spin rate was increased to 2500 rev min$^{-1}$, better film uniformity owing to reduction in island effect and better film lustre were obtained but, the thickness falls. This may be attributed to the enhanced outward flow and uniform thinning of the sol in the coating. Less island formation (i.e. better film uniformity) may tend to decrease the sheet resistance whereas thickness reduction will tend to increase the sheet resistance. When the turntable spin rate is increased to 3000 rev min$^{-1}$, the contribution from the first effect may be a little dominant resulting in lowering of the resistivity of the film. At 3500 rev min$^{-1}$ the film uniformity has been
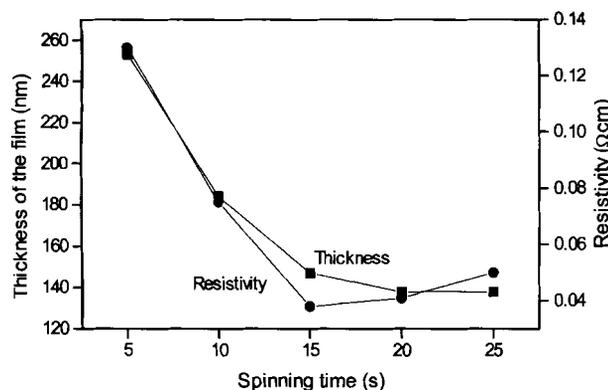


**4 Variation of resistivity and thickness of TO films with spinning time $t$**
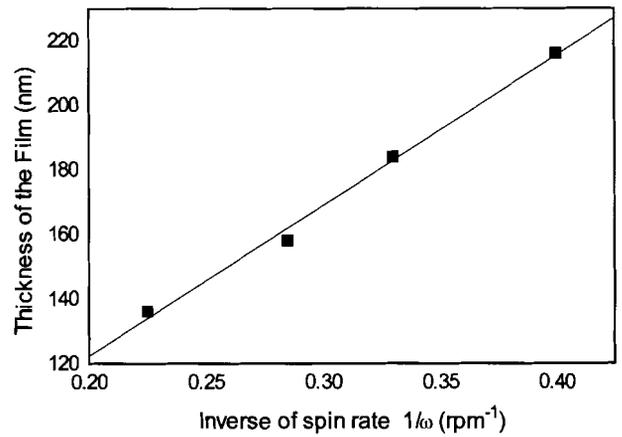


**5 Variation of thickness with inverse of spin rate of TO films**

found optimum without any island formation and at which stage the resistivity is also minimum. At spin rates above 3500 rev min$^{-1}$ most quantity of the sol is thrown out from the surface owing to the action of high degree of centrifugal force. At this point, the solvent evaporation mechanism and shear rate variation dominate, which reduces the film thickness. So the contribution from the second factor mentioned earlier may dominate resulting in a relatively higher resistivity of the film. Hence 3500 rev min$^{-1}$ has been selected as the turntable spin rate.

## Fixing turntable spin time

In the spin coating technique, the turntable spin time is a parameter to be fixed for obtaining uniform good films. To optimise the time of spinning, the spinning speed is fixed at 3500 rev min$^{-1}$ and the heat treatment temperature at 400°C, the heat treatment time at 5 min and the number of coatings at 6. The variation of resistivity of the films with spin time is given in Fig. 4.

Initially when 5 s was used as the spin time, films of poor lustre and uneven surfaces with islands of yellowish white patches and a consequent high resistivity were obtained. As the time of spinning was increased the lustre, the surface smoothness and film uniformity improved and the resistivity of the film decreased and at 15 s of spinning time good film with minimum resistivity was obtained. Further increase in the time of spinning resulted in a marginal increase in resistivity. The plot of film thickness with spinning time shows that initially, i.e. at $t=5$ s, the film thickness is relatively high and it decreases on increasing the time of spinning and this happens up to 15 s beyond which the thickness variation is rather small reflecting the behaviour exhibited by resistivity. So any turntable spin time between 15 and 25 s is suitable and for the present study 15 s has been fixed as the turntable spin time. The variation of the film thickness $h$ with spin time $t$ is shown in Fig. 4.

A simple theoretical model has been proposed by Hirasawa[22] and Doughton and Given[23], in this they arrived at the equation for film thickness $h$ as

$$h = \frac{h_o}{\left(1 + \frac{4\rho\omega^2}{\eta}h_o^2 t\right)^{1/2}} \qquad (1)$$
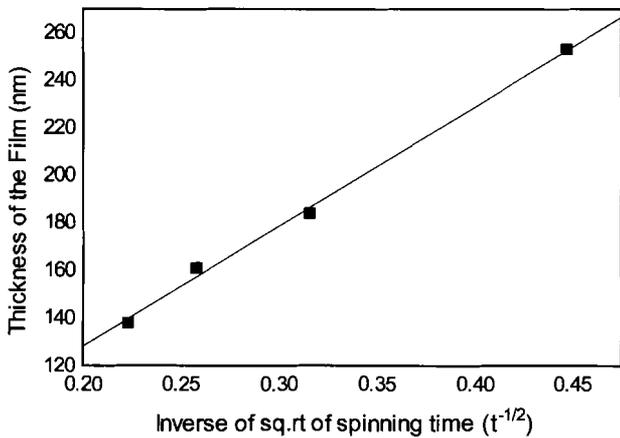
**6 Variation of thickness with square root of spinning time of TO films**

where $h_o$ is the value of $h$ at $t=0$, $\rho$ the density of the liquid, $\omega$ the spin rate and $\eta$ the coefficient of viscosity.

From the equation it is seen that

$$h\alpha(1/\omega) \text{ and } h\alpha(1/t^{1/2})$$

Following Peuruung and Graves,[24] if it is assumed that drying/heat treatment has no significant effect on the shape of the film profile, $h\alpha(1/\omega)$ and $h\alpha(1/t^{1/2})$ may also be taken to be reasonably valid for dried/heat treated films. A plot (Fig. 5) between experimental values of the film thickness $h$ and reciprocal of the spin rate $1/\omega$ is more or less a straight line indicating that $h$ is nearly proportional to $1/\omega$. Also the near straight line plot (Fig. 6) between the film thickness $h$ and the reciprocal of square root of time of spinning confirms that $h$ is proportional to $1/t^{1/2}$. Thus, the experimental results of the present study agree with the theoretical prediction of Doughton and Given[23] and Hirasawa et al.[22]

The dependence of the thickness of sol–gel spin coated $SnO_2$ films on the number of coatings is shown in Fig. 7. The variation is found to be more or less linear and practically no detectable change in film thickness has been observed in the heat treatment temperature range 375–425°C. This conclusion is consistent with that arrived at by Lee et al.[25] while studying the thickness variation of sol–gel derived $SnO_2$ films on the number of coatings. The average film thickness/coat is found to be 25 nm which is nearly the same as that reported by Cobianu et al.[17]
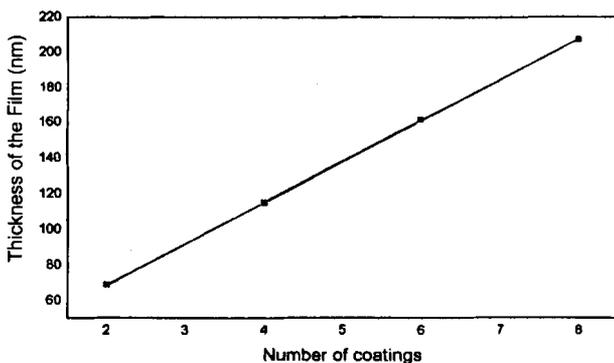


**7 Variation of thickness of TO films with number of coatings**
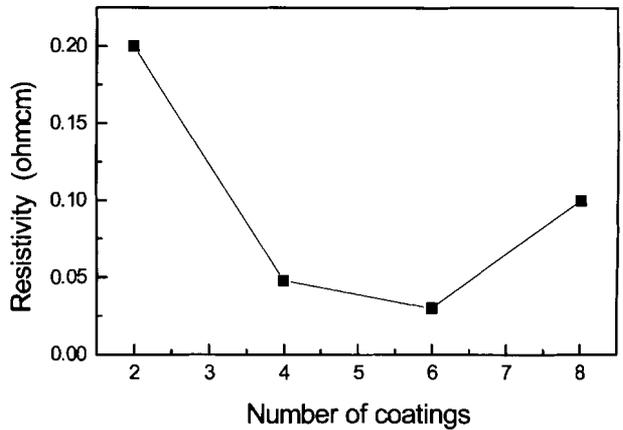


**8 Variation of resistivity of TO films with number of coatings**

## Optimisation of number of coatings

Keeping the heat treatment temperature at 400°C, the number of coatings has been varied and the variation of resistivity of the tin oxide films with number of coatings is shown in Fig. 8. The number of coatings decides the thickness of the film – the greater the number of coatings, the larger the thickness of the film. From the Fig. 8, it is seen that as the number of coatings increases from a low value, i.e. as the film thickness increases, the resistivity of TO film decreases, reaches a minimum value at 6 coatings and then increases. This result is rather intriguing. But this can be qualitatively understood if the factors contributing to the conductivity of the film are considered.

When the number of coatings is small, crystallinity and size of crystallites may be low and the surface scattering may be considerable owing to small mean free path, giving rise to large grain boundary scattering and these would be contributing to high resistivity. At the same time, since the number of heat treatment steps is small, the oxygen vacancies contribute to the conductivity tending to lower the resistivity. Since the former factor may be dominant over the latter factor the film would show high resistivity only. When the number of coatings increases the film thickness increases, the surface scattering may be reduced owing to increased mean free path and the crystallinity and size of the crystallite may improve tending to improve the conductivity. But as the number of heat treatment steps is to be consequently increased, the number of oxygen vacancies may be lowered tending to rise the resistivity. Under the influence of these two, the resistivity of the film falls because of the dominance of the first factor. This fall will continue to occur as long as the first effect dominates. After a number of coatings and consequent number of heat treatment steps, the oxygen vacancies may become very low resulting in further increase in resistivity and this may become dominant over the reduction in resistivity owing to the first factor mentioned above. Therefore the resistivity of the films may increase. The minimum resistivity occurs at the sixth coating. Thus the optimum number of coatings is taken as 6 for the tin oxide films.

## Optimisation of heat treatment temperature

Heat treatment temperature is one of the parameters, which may influence the stoichiometry and structural
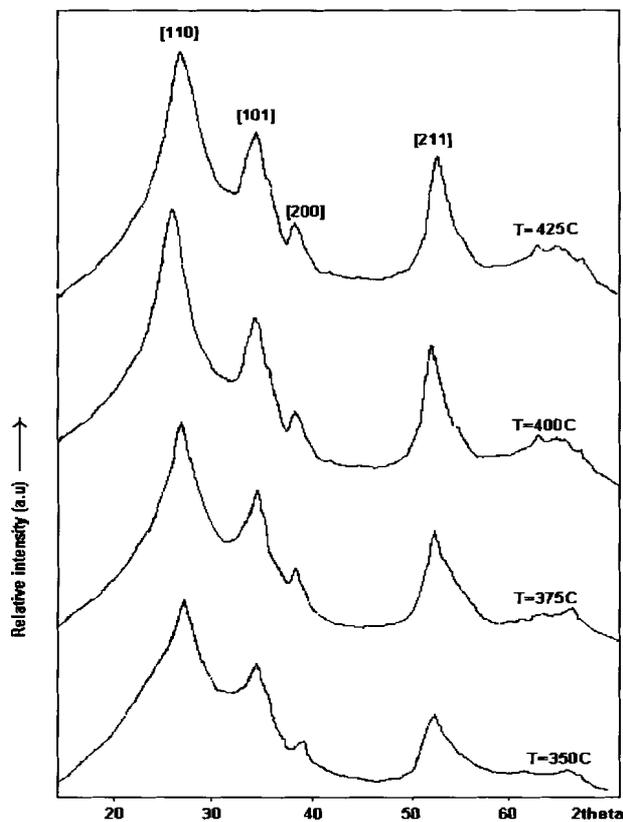
9 XRD patterns of tin oxide films formed at 350, 375 and 425°C



10 Transmittance spectra of TO thin films developed at 400°C with various numbers of coatings

properties of the films. The Fig. 9 shows the XRD of TO films prepared at different heat treatment temperatures varying from 350 to 425°C. From the figure, it is seen that the films are polycrystalline in nature with preferred growth direction along [110]. Other peaks corresponding to the directions [101], [200] and [211] are also seen. All the diffraction patterns shown in the Fig. 9 are characteristic of TO structure (tetragonal rutile structure). The larger values of full width at half maximum (FWHM) for all peaks at all temperatures suggest the formation of naoncrystalline $SnO_2$. It is observed from the figure that as the heat treatment temperature increases the intensity of the preferred orientation [110] also increases, indicating an improved crystallinity and crystallite size. This is reflected by other peaks, too; for example, at 350°C the [200] peak is very weak, but it shows up as a well marked peak at 400°C and 425°C. The intensity of the [211] peak increases with temperature. Shanon et al.[3] have also observed that the [200] peak gets intensified at higher temperatures, while analysing the TO films developed by chemical vapour

deposition (CVD). However, no phase change at elevated temperatures as reported[2] to occur in TO films has been noticed in the spin coated films with heat treatment temperatures in the range 350–425°C.

Thus it can be seen that the intensity of the peaks obtained increases with increase in heat treatment temperature indicating an improved crystallinity and crystallite size with temperature. This result is consistent with the conclusion arrived at by Vasu and Subrahmanyam[13] and Shanon et al.[3] using other film preparation techniques.

Regular, strong and sharp crystalline peaks are found even at a relatively low temperature of 350°C itself. Contrastingly, Lin and Wu[2] who have grown TO films with gel suspension on glass substrates could not get distinguishable crystalline peaks at 300°C and only at a much higher temperature of 500°C, they could obtain crystalline peaks.

The XRD data for the TO films developed at various heat treatment temperature are presented in Table 1. The data agree fairly well with the values given in Joint Committee for Powder Diffraction Standards (JCPDS) data sheets. The calculated values of lattice parameters $a=4\cdot73$ Å, $c=3\cdot19$ Å are in very good agreement with the values of $a=4\cdot737$ Å, $c=3\cdot185$ Å reported by Binoy.[26]

It is found that the average grain size varies from 1·92 to 4·80 nm as the heat treatment temperature varied from 350 to 425°C. This result confirms the formation of nanocrystalline $SnO_2$ films by the sol–gel spin coating technique at relatively low temperatures.

## Optical and electrical properties

The transmittance spectra of these films are shown in Fig. 10 and the optical data are presented in Table 2. It is seen that as the number of coatings increases, the

Table 1 XRD data for tin oxide films with different heat treatment temperature (six coatings)

| Standard data | | | Heat treatment temperature, °C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| JCPDS file no. 6–416 | | | 350 | | | 375 | | | 400 | | | 425 | | |
| hkl | d, Å | I/I₀ | 2θ | d, Å | Peak count | 2θ | d, Å | Peak count | 2θ | d, Å | Peak count | 2θ | d, Å | Peak count |
| 110 | 3·347 | 100 | 26·5 | 3·351 | 1395 | 26·6 | 3·351 | 1397 | 26·6 | 3·351 | 1425 | 26·6 | 3·351 | 1439 |
| 101 | 2·643 | 75 | 33·7 | 2·651 | 1044 | 33·7 | 2·643 | 1069 | 33·7 | 2·643 | 1069 | 34·1 | 2·652 | 1093 |
| 200 | 2·369 | 21 | 37·4 | 2·387 | 604 | 37·9 | 2·380 | 640 | 37·9 | 2·380 | 640 | 37·8 | 2·380 | 629 |
| 211 | 1·764 | 57 | 51·8 | 1·767 | 697 | 51·9 | 1·765 | 921 | 51·9 | 1·765 | 921 | 51·9 | 1·765 | 823 |

12 Transmittance spectra of TO films developed at 350, 375, 400 and 424°C

11 $\alpha^2$ versus $h\nu$ plots of TO films with two, four, six and eight coatings

absorption edge shifts to the lower wavelength side and is comparatively low when the number of coatings is 4–6. The transmittance at 550 nm is high when the number of coatings is small and gets lowered and becomes more or less constant when the number of coatings is between 4 and 8. The film porosity, surface and structural homogeneity and crystallinity are factors that have positive influence on the transmittance of transparent conducting oxide (TCO) films.[27,28] When the number of coatings is small, the film thickness is low and there may be less uniformity (i.e. greater porosity) which may tend to increase the transmittance.

The surface and structural homogeneity may be less and the crystallinity and size of the crystallites may be low which may tend to decrease the transmittance. The

former effect may be dominant when the film thickness is low and this may result in relatively higher transmittance. This is consistent with the contention of other authors[27,28] that low film thickness leads to high transmittance. When the number of coatings increases, i.e. when the film thickness increases, the porosity would decrease, i.e. film uniformity will improve, tending to decrease the transmittance; the surface and structural homogeneity and crystallinity and size of crystallites may improve tending to increase the transmittance.

Under these effects, the transmittance may fall and may become more or less constant beyond a thickness, i.e. beyond a certain number of coatings. Such near constancy of transmittance at higher film thickness has been reported.[26] The band gap values from the $\alpha^2$ versus $h\nu$ plot (Fig. 11) show that the band gap energy is 3·46 eV and does not vary much with the number of coatings, i.e. with the film thickness. The transmittance spectra of the TO films developed at 350, 375, 400 and 425°C are shown in Fig. 12 and the spectral data in Table 3.

The transmittance marginally increases from 350 to 375°C and becomes more or less constant with a value ~93·6%. Such a trend has been observed by Lin and Wu[2] also, for their colloidal gel suspension derived $SnO_2$ films. The absorption edge shifts to lower wavelength side as the heat treatment temperature increases and is minimum (300 nm) at 400°C at which stage the band edge is 3·46 eV (Fig. 13).

The linearity of the $(\alpha h\nu)^2$ versus $h\nu$ plot (Fig. 14) for the $SnO_2$ film, prepared under the above mentioned optimised conditions, indicates that the absorption near

Table 2 Optical properties of tin oxide films at different number of coatings at heat treatment temperature 400°C

| No. of coatings | Thickness, nm | Absorption edge, nm | Transmittance at 550°C, % | Band gap, eV |
|---|---|---|---|---|
| 2 | 69 | 311 | 97·0 | 3·43 |
| 4 | 115 | 300 | 91·2 | 3·46 |
| 6 | 161 | 300 | 93·6 | 3·46 |
| 8 | 207 | 311 | 91·5 | 3·44 |

Table 3 Optical properties of tin oxide films at different heat treatment temperatures six coatings

| Temp., °C | Thickness, nm | Absorption edge, nm | Transmittance at 550 nm, % | Band gap, eV |
|---|---|---|---|---|
| 350 | 161 | 318 | 89·9 | 3·39 |
| 375 | 161 | 314 | 94·5 | 3·41 |
| 400 | 161 | 300 | 93·6 | 3·46 |
| 425 | 161 | 318 | 93·2 | 3·42 |
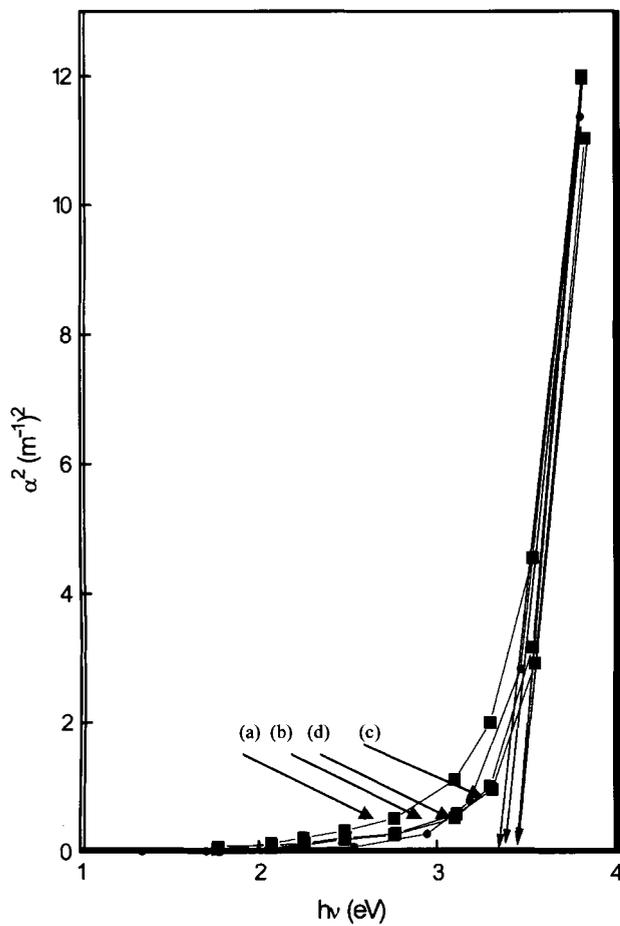
**13** $\alpha^2$ versus $h\nu$ plots of TO films found at 350, 375, 400 and 425°C
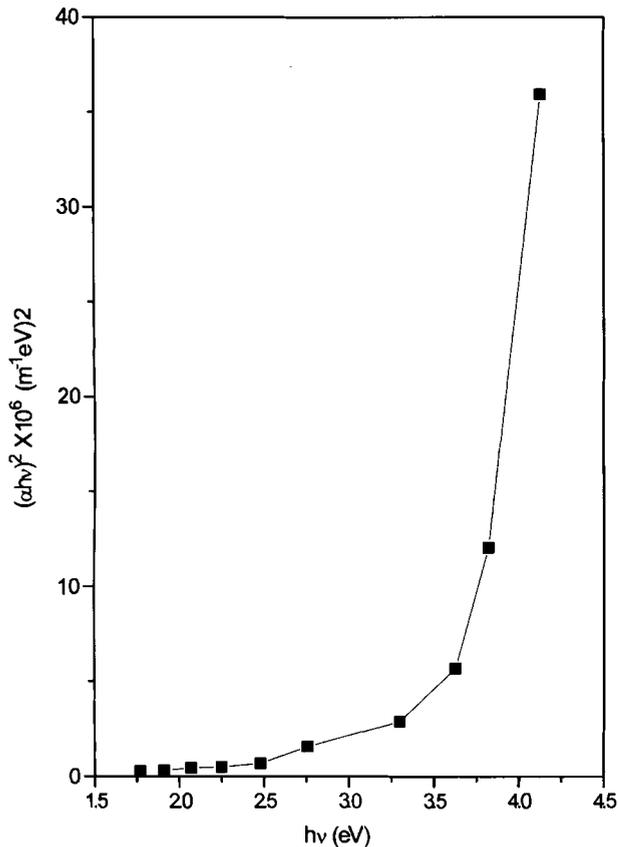


**14** $h\nu$ versus $(\alpha h\nu)^2$ plot for tin oxide film formed at the optimised conditions
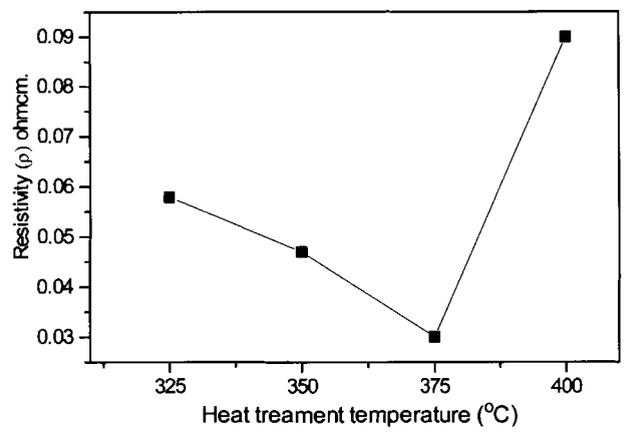


**15** Variation of resistivity of tin oxide thin films with heat treatment temperature
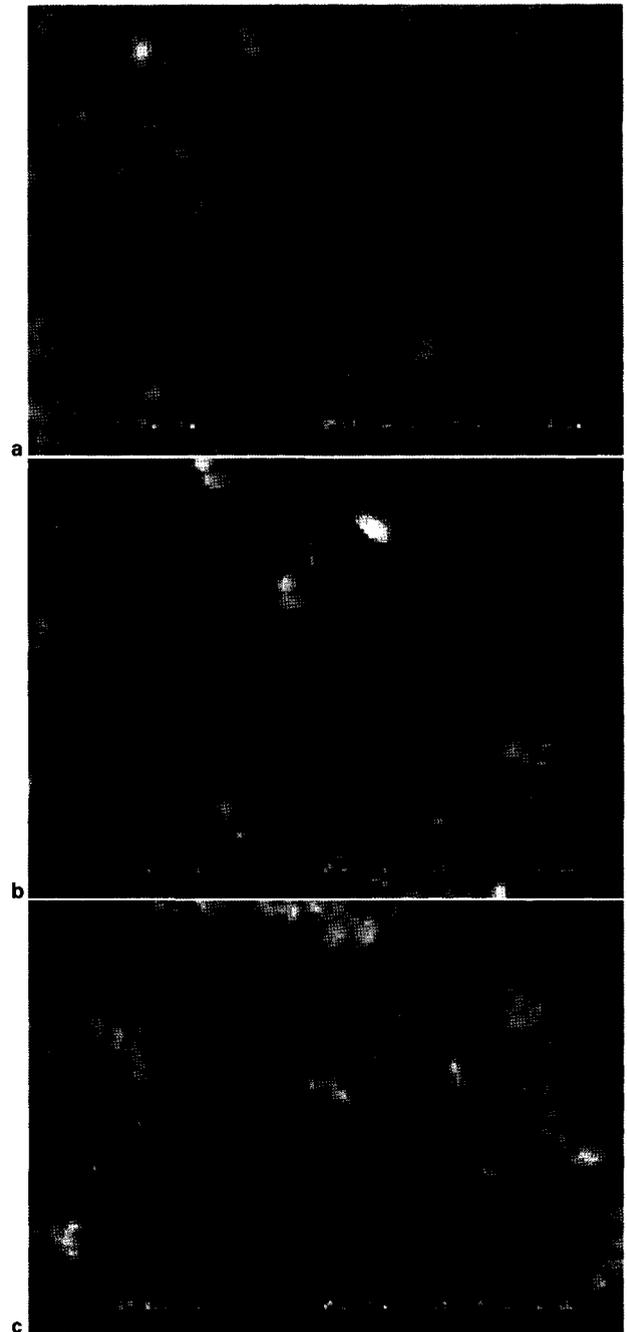


**16** SEM images of TO films prepared at *a* 375, *b* 400 and *c* 425°C
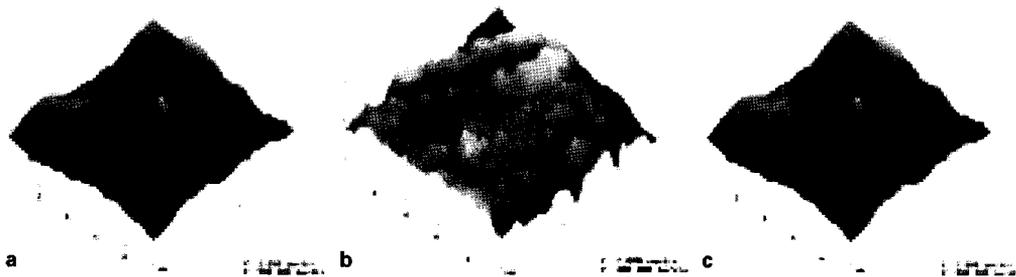
**17 AFM images of TO films prepared at *a* 375, *b* 400 and *c* 425°C**

the fundamental edge is due to direct allowed transition. Further the value of $\alpha$ is of the order of $10^4$ cm$^{-1}$ which is characteristics of direct allowed transition.[28] At the optimum heat treatment temperature of 400°C with 6 coatings, the transmittance is 93·6% with a band gap of 3·46 eV and at this stage the resistivity is also minimum at $3·0 \times 10^{-2}$ $\Omega$ cm. This resistivity value is in the range of values reported for TO films grown under optimised conditions by Uen *et al.*[8] ($\rho$ ranging from $7·2 \times 10^{-2}$ to $2·0 \times 10^{-2}$ $\Omega$ cm) at $T_{sub}$=400°C using the reactive evaporation method (with a dc glow discharge of oxygen); Reddy *et al.*[18] have obtained $\rho$=9·0 $\times$ $10^{-2}$ $\Omega$ cm at an annealing temperature of 500°C for the film prepared using electron beam deposition technique, Vasu and Subrahmanyam[13] ($\rho$ ranging from $2·1 \times 10^{-3}$ to $1·4 \times 10^{-2}$ $\Omega$ cm, $T_{sub}$=280–440°C, $E_g$= 3·76–3·86 eV) using spray pyrolysis method, Shanon *et al.*[3] ($\rho$ ranging from $4·0 \times 10^{-3}$ to $1·0 \times 10^{-2}$ $\Omega$ cm, $T_{sub}$=300–370°C) using the CVD.

The variation of resistivity of TO films with respect to the heat treatment temperature is shown in Fig. 15. The plot shows that as the temperature increases from 350°C the resistivity falls and is minimum at 400°C and then rises. As the heat treatment temperature rises, the crystallinity and size of the crystallites may improve, the boundary and surface scattering may be reduced all tending to improve the film conductivity; but the number of oxygen vacancies will decrease tending to decrease the conductivity. If the first factor dominates, the conductivity will be high (and the resistivity will be low). This seems to happen at and below 400°C for sol–gel spin coated $SnO_2$ films. However, if the second influence dominates over the first, the conductivity will decrease and the resistivty will increase. This happens in the present case at 425°C. A minimum resistivity of $3·0 \times 10^{-2}$ $\Omega$ cm (sheet resistance 528 $\Omega$/square) has been obtained. Contrastingly Lin and Wu[2] obtained an optimised sheet resistance of $5 \times 10^7$ $\Omega$/square for their sol–gel suspension deposited TO films at 500°C. The present study shows that good quality, TO films at a relatively low heat treatment temperature of 400°C and low heat treatment time of 10 min can be obtained using the spin coating technique.

## Surface morphological studies

Analyses of the surface morphology by SEM studies of the spin coated $SnO_2$ films prepared at temperatures 375, 400, 425°C have been conducted and the micrographs are shown in Fig. 16*a, b* and *c* respectively. $SnO_2$ films prepared at 375°C show a uniform surface with few pin holes and patches. At 425°C (Fig. 16*c*) there are more agglomerations seen all over the surfaces. Fig. 16*b* shows a uniform surface morphology for the $SnO_2$ film

prepared under the optimised conditions at 400°C. Below and above this optimum temperature the surface morphology is not as good as that of the film prepared at 400°C.

The AFM pictures are shown in Fig. 17 for the $SnO_2$ films prepared at 375°C, 400°C and 425°C. The present study also reveals the uniformity of surface with grain size of ~0·5 $\mu$m. This is in support of the SEM surface morphology observed for these films. $SnO_2$ film prepared at 375°C shows some non-uniform surface morphology. As heat treatment temperature increases to 400°C the surface morphology becomes smooth and more uniform. But, when the temperature of heating the film is 425°C the surface morphology once again becomes less uniform. These surface morphology studies show that the spin coating technique can be used for the preparation of large area tin oxide films at a temperature of 400°C for solar cells and other device applications.

## Conclusion

The present study clearly demonstrates that device quality, smooth and uniform $SnO_2$ films with good lustre and with a resistivity of $3·0 \times 10^{-2}$ $\Omega$ cm and a band gap of 3·46 eV have been developed by the spin coating method at a relatively low heat treatment temperature of 400°C and relatively small heat treatment time of 5 min/coat.

## References

1. M. I. B. Benardi, C. M. Barrado, L. E. B. Soledade, E. R. Leite, E. Longo and J. A. Varela: *J. Mater. Sci. Lett.*, 2002, **21**, 1241.
2. Y. J. Lin and C. J. Wu: *Surf. Coat. Technol.*, 1996, **88**, 239.
3. G. Shanon, Raj Rup and A. Manansingh: *Thin Solid Film.*, 1990, **190**, 287.
4. J. Jeong, S. P. Choi, C. I. Chang, D. C. Shin, J. S. Park, B. T. Lee, Y. J. Park and H. J. Song: *Solid State Commun.*, 2003, **127**, 595.
5. Y. I. Kim, S. H. Nahm and M. J. Jung: *Mater. Lett.*, 2003, **57**, 3653.
6. A. M. Serventi, D. G. Rickerby, M. C. Horrillo and R. G. S. Jacques: *Thin Solid Film.*, 2003, **445**, 38.
7. B. S. Chiou, S. T. Hsieh and W. F. Wu: *J. Am. Ceram. Soc.*, 1994, 77, 1740.
8. T. M. Uen, K. F. Huang, M. S. Chen and Y. S. Gou: *Thin Solid Films*, 1988, **158**, 69.
9. R. P. Howson, H. Barankova and A. G. Spenoz: *Thin Solid Films*, 1991, **196**, 315.
10. B. K. Min and S. D. Choi: *Sens. Actuators B*, 2004, **98**, 239.
11. B. P. Kryzhanovsku and E. N. Ore: *Sov. J. Opt. Technol.*, 1974, **41**, 479.
12. S. Muranaka, Y. Bando and T. Takada: *Thin Solid Film.*, 1981, **86**, 11.
13. V. Vasu and S. Subrahmanyam: *Thin Solid Film.*, 1990, **189**, 217.
14. A. Tucic, Z. V. Marinkovic, L. Mancic, M. Cilense and O. Miloevic: *J. Mater. Proc. Technol.*, 2003, **143–144**, 41.
15. G. Korotcenkov, V. Brinzari, V. Golovanov and Y. Blinov: *Sens. Actuators B*, 2004, **98**, 41.

16. S. Ray, R. Banerjee, N. Basu, A. K. Batabyal and A. K. Barua: *J. Appl. Phys.*, 1983, **54**, 3497.
17. C. Cobianu, C. Savaniu, O. Buiu, D. Dascalu, M. Zaharescu, C. Parlog, A. Berg and B. Pecz: *Sens. Actuators B*, 1997, 43, 114.
18. M. H. M. Reddy, S. R. Jawalekar and A. N. Chandorkar: *Thin Solid Film.*, 1990, **187**, 171.
19. R. S. Niranjan and I. S. Mulla: *Mater. Sci. Eng. B*, 2003, **103**,103.
20. B. Esfandyarpour, S. Mohajerzadeh, S. Famini, A. Khodadadi and E. A. Soleimani: *Sens. Actuators B*, 2004, **100**, 190.
21. F. Gu, S. F. Wang, M. K. Lü, X. F. Cheng, S. W. Liu, G. J. Zhou, D. Xu and D. R. Yuan: *J. Cryst. Growth*, 2004, **262**, 182.
22. S. Hirasawa, Y. Saito, H. Nezu, N. Ohashi and H. Maruyama: *IEEE Trans. Semicon. Manuf.*, 1997, **4**, 438.
23. W. J. Daughton and F. L. Givens: *J. Electrochem. Soc. Solid State Sci. Technol.*, 1985, **129**, 173.
24. L. M. Peuruung and D. B. Graves: *IEEE Trans. Semiconduct. Manufact.*, 1993. **6**, 72.
25. S. C. Lee, J. H. Lee, T. S. Oh and Y. H. Kim: *Sol. Energ. Mater. Sol. Cells*, 2003, **75**, 481.
26. M. D. Binoy: PhD thesis, An investigation on the preparation of some transparent conducting oxides and lead selenide thin films, Cochin University of Science and Technology, Cochin, Kerala, India, 2001.
27. S. S. Kim, S. Y. Choi, C. G. Park and H. W. Jin: *Thin Solid Film.*, 1999, **347**, 155.
28. E. Pommier, C. Gril and J. Maruchi: *Thin Solid Film.*, 1981, **77**, 91.