Indium Zinc Oxide as a Transparent Conductor for

Thin Film Solar Cell Applications

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Introduction

Transparent conducting oxides (TCO) films have gained significant importance due to their low resistivity and high transparency. They are largely used in devices such as flat panel displays and thin film solar cells such as a-Si, CIGS, CdTe, and CZTS.

Traditionally, many of the thin-film solar cells used indium tin oxide (ITO) as the TCO. In recent years, due to unstable pricing and a relatively small indium supply, new materials have been sought to either reduce or remove indium. So far a good progress is ZnO doped with aluminum. But the Al doped ZnO films suffer from loss in transmission due to high carrier concentrations particularly in the near-infrared range.

A much less investigated area in the ZnO-In_2O_3 material system is when the Zn/(Zn+In) ratio is greater than 0.5 and less than 0.9 which consists of the $Zn_xIn_2O_{x+3}$ type structure. Indium presented in this type of structure may allow for enhanced mobility without increasing carrier concentration and thus to yield films with better transmission and with good conducting properties.

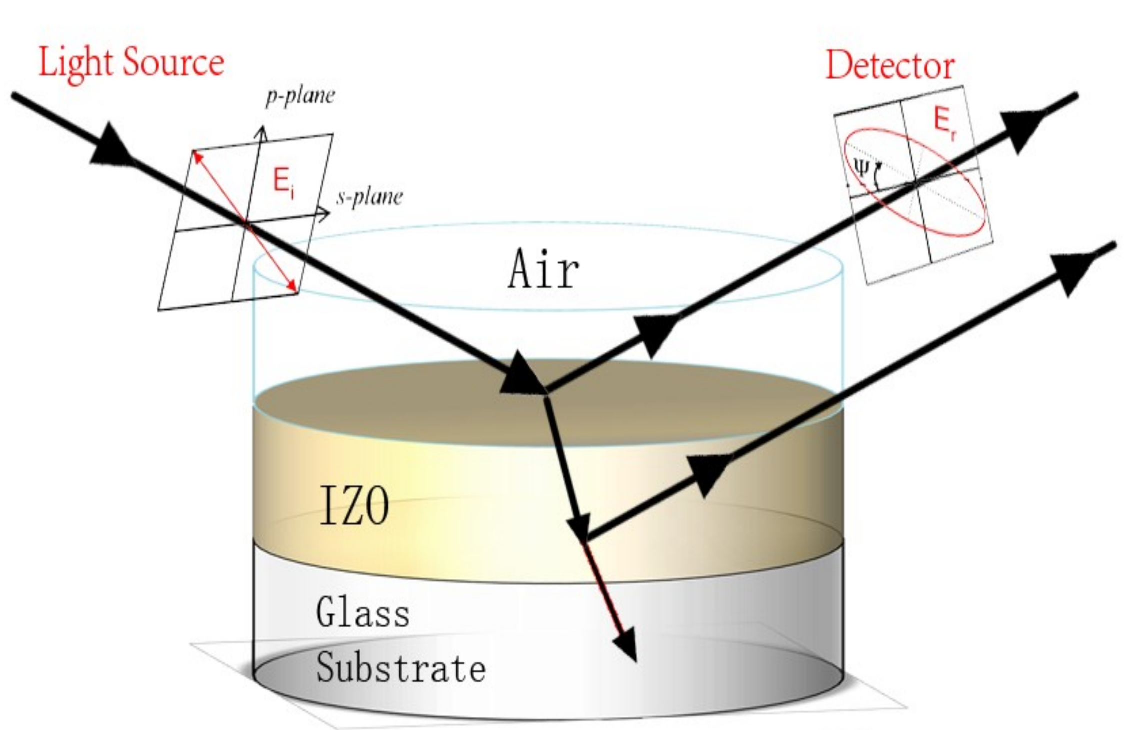
In this study, ZnO and pure Indium targets were co-sputtered to form InZnO by varying oxygen concentration in mixture of Argon/Oxygen and under different DC sputtering powers to the Indium target. Prepared films were characterized with UV-Vis-NIR spectroscopic ellipsometer and reflectometers. Electrical properties were also evaluated.

Experimental

All films were produced with a magnetron sputtering system. Three 3" sputtering guns were used. Two RF guns for ZnO and one DC gun for pure Indium target was installed on the sputtering system. Soda-lime glass (SLG) was used as substrates. Samples were deposited at room temperature. Chamber pressure was maintained at 2mT. The deposition power for ZnO was kept at 120 watts, while the deposition powers for indium were set at three different power levels: 20, 40, and 60 watts. During deposition, total gas flow rate at 10 sccm was kept constant, but oxygen content in Argon-Oxygen mixture varied from 0 to 4% to investigate the oxygen effect.

Film Characterization

The film thickness and optical constants of the InZnO films were characterized using a spectroscopic ellipsometer TFProbe SE500BA, developed by Angstrom Sun Technologies Inc. TFProbe SE500BA has a wavelength range from 250 to 1700nm with a resolution better than 1nm for UV-Visible and 3nm for near-infrared range. This ellipsometer is also equipped with a high precision Goniometer for automatic variable angle measurement. All data were acquired at 65, 70 and 75 degree three incident angles with 512 wavelength points. The physical and optical properties of the InZnO films was obtained by fitting the measured ellipsometry parameters, Psi (Ψ) and Delta (Δ), to the model data obtained with the theoretically calculated set of parameters.



Using the TFProbe 3.3 software, a Tauc-Lorentz dispersion model was applied to obtain the optical functions of the InZnO films. The imaginary part of such dielectric function ε_i was developed by Jellison and Modine in 1996, by multiplying the Tauc joint density of states with the Lorentz oscillator:

$$\varepsilon_i(E) = \frac{AE_0C(E-E_G)^2}{E\left((E^2-E_0^2)^2+C^2E^2\right)}, \quad E > E_g$$

$$\varepsilon_i(E) = 0, \quad E \le E_g$$

where E_0 is the peak transition energy, C is the broadening term, E_g is the optical band gap, and A is proportional to the transition probability matrix element.

The real part of the dielectric function ε_r is calculated then by Kramers-Kronig integration:

$$\varepsilon_r(E) = E_{inf} + \frac{2P}{\pi}\int_{E_g}^{\infty}\frac{\xi\,\varepsilon_i(\xi)}{\xi^2-E^2}\,d\xi$$

The fitting parameters in the software utilizes the variables A, C, E_0, E_g and E_{inf} from the Tauc-Lorentz model.

When studying optical property over a very wide wavelength range, especially for TCO films, a Tauc-Lorentz dispersion is insufficient to describe the dielectric response completely. Therefore, one or more Lorentz type oscillators were added into the total dielectric function in the analysis

$$\varepsilon_r = \frac{A_1\lambda^2(\lambda^2-L_0^2)}{(\lambda^2-L_0^2)^2+\gamma^2\lambda^2} \qquad \varepsilon_i = \frac{A_1\lambda^3\gamma}{(\lambda^2-L_0^2)^2+\gamma^2\lambda^2}$$

where A_1 is the amplitude, L_o is the central wavelength and γ is the width of the oscillators.

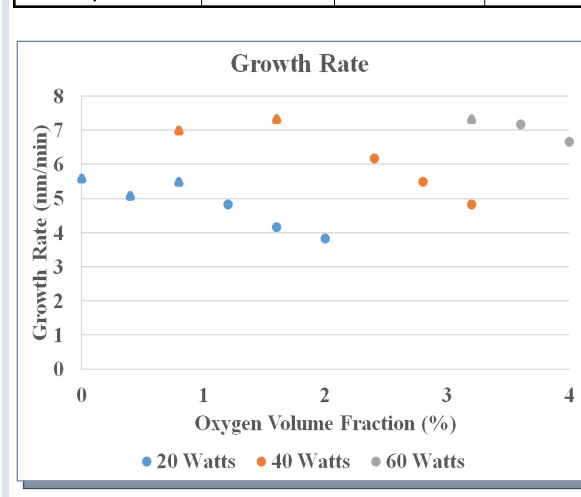
The Levenberg-Marquardt algorithm (LMA), a non-linear least-Squares method, is used for modeling. The best fitted variables are found by minimizing the difference between the measured and model data.

TFProbe SR500RT spectroscopic reflectometer was used for sample performance evaluation by measuring each sample's reflection and transmission spectra. In addition, 1cm^2 square samples, cleaved from the glass slides, were used for electrical measurements with a four point probe. Four indium dots were soldered onto four corners for hall measurements. X-ray photoelectric spectroscopy (XPS) technique was used to

Results

Film Thickness and Growth Rate

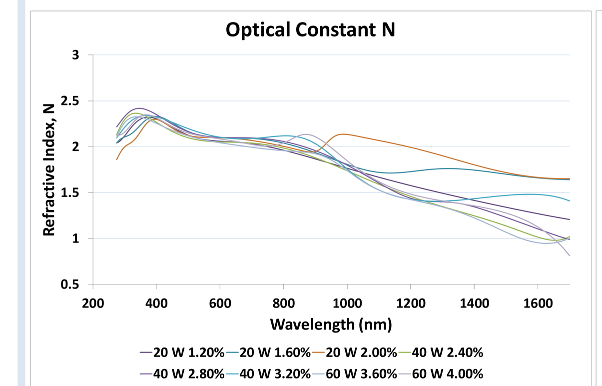
Sample Information (Indium power, Oxygen %)	Thickness Results from TFProbe Spectroscopic Ellipsometer SE500BA					Film Thickness from Profilometer (nm)
	Roughness (nm)	Uncertainty	Thickness (nm)	Uncertainty	Total Thickness (nm)	
20 W, 1.20%	5.95	± 0.90	294.13	± 2.05	300.08	290
20 W, 1.60%	8.95	± 1.10	253.86	± 1.64	262.82	250
20 W, 2.00%	9.14	± 1.29	209.15	± 1.64	218.29	230
40 W, 2.80%	6.23	± 1.47	361.89	± 2.36	368.11	330
40 W, 3.20%	10.08	± 0.81	327.96	± 1.44	338.03	290
60 W, 3.60%	9.21	± 1.20	411.55	± 2.84	420.76	430
60 W, 4.00%	5.51	± 1.17	425.45	± 2.75	430.96	440

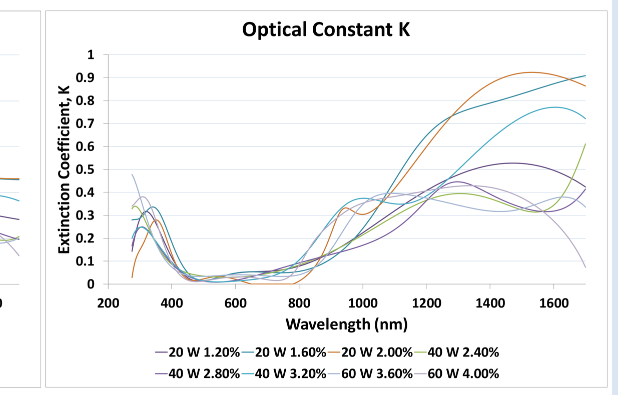


To achieve the best fitting, a roughness layer was added to the ellipsometry model. In the table above, a total thickness was summed for each sample. Physical thickness was measured with an alpha-step profilometer is also listed in the same table, which illustrated both results are consistent. Calculated growth rates show (a) the film growth rate decreases when oxygen content increases at the same DC power for indium target, (b) the growth rate increases when sputtering power increases at the same level of oxygen volume fraction in gas mixture.

Optical Constants

Optical constants, refractive index (N) and extinction coefficient (K), were plotted in the figures below for select samples. From the K plot, these selected samples exhibit typical characteristics of TCO film with low absorption in the visible wavelength range.



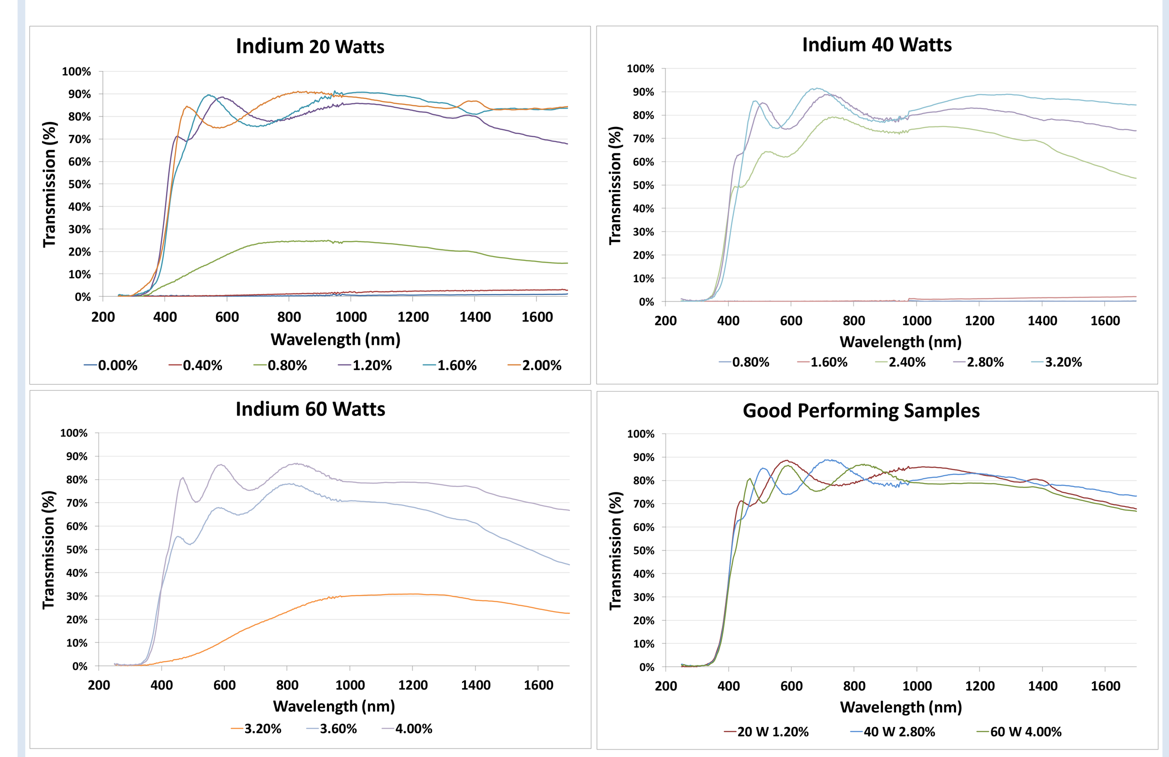


Band Gap

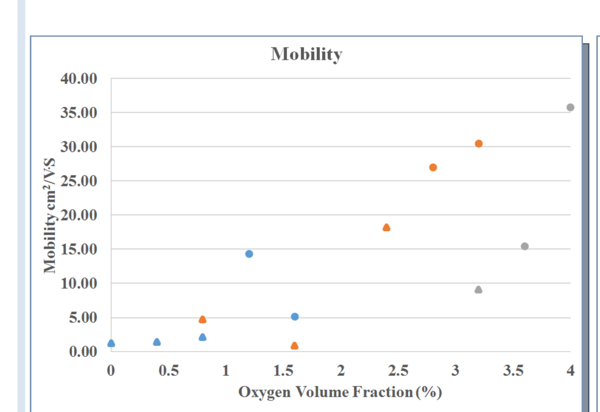
Optical band gap (E_g), obtained through the Tauc-Lorentz dispersion model, are ranged from 2.6eV to 3.2eV, which are in the same range as the estimated Tauc plot obtained with the transmission spectra.

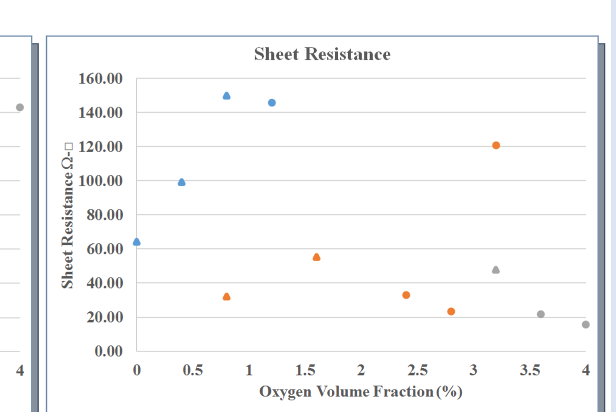
Sample Information (Indium power, Oxygen %)	Tauc-Lorentz Parameters				
	A	C	E_0	E_g	E_∞
20 W, 1.20%	537.06	1.3098	2.6211	**2.7987**	2.0168
20 W, 1.60%	520.76	1.3291	2.6581	**2.7064**	2.2048
20 W, 2.00%	560.19	1.0857	2.1210	**2.6807**	2.6393
40 W, 2.40%	168.86	2.4440	4.8748	**2.6288**	2.3969
40 W, 2.80%	153.90	4.5723	9.1444	**2.6979**	1.0664
40 W, 3.20%	148.31	4.6316	9.2630	**2.6113**	1.1839
60 W, 3.60%	237.14	5.4062	11.3455	**3.1214**	3.3896
60 W, 3.63%	182.12	2.4242	4.9055	**2.9128**	3.3165
60 W, 4.00%	125.77	2.4404	4.9891	**2.6629**	4.1219

Transmission Spectra



Electrical Properties





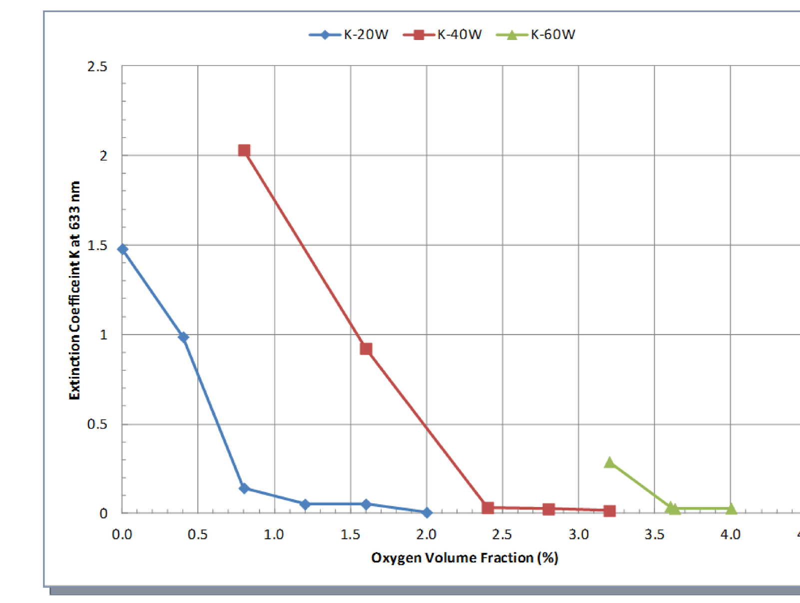
From the mobility data, higher oxygen is needed to obtain higher mobility. Resistance measurement illustrates medium to high power levels for indium target gives a smaller resistance trend. Good performance can be seen with processing condition at 40w power with 2.8% oxygen content.

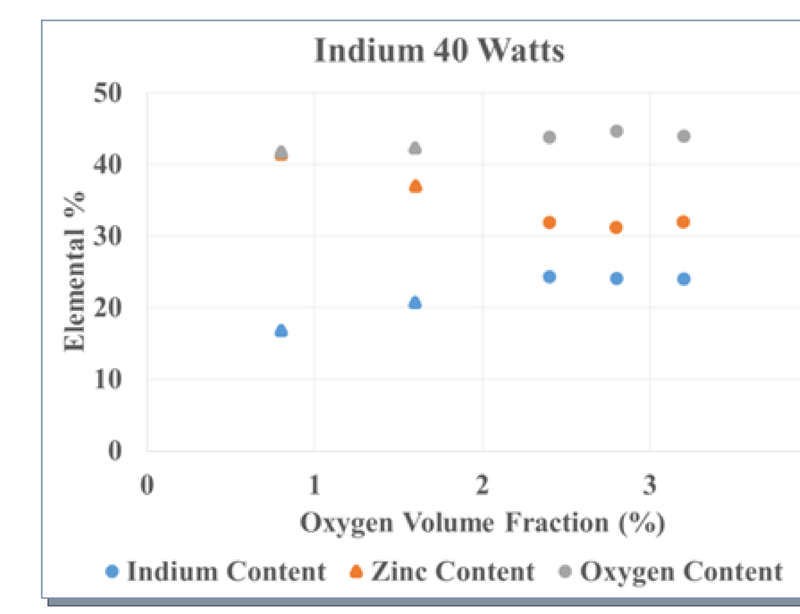
Discussion and Summary

The effect of the processing parameters to absorption behavior of the films, extinction coefficients K at a wavelength of 633 nm, was plotted against volume fraction of Oxygen at three different levels of sputtering power for indium target.

(1) Absorption of films decreases with the increase of oxygen concentration in argon-oxygen mixture at each sputtering power

(2) Relatively higher oxygen concentration is needed to achieve smaller absorption at higher sputtering power.





By reviewing the electrical properties, optical properties and analyzed composition of samples in this preliminary study, 40 watts DC power with higher oxygen volume fraction at 2.5% to 3% is the ideal processing condition to achieve a good performance TCO film.

The InZnO film was deposited through a relatively simple deposition at room temperature without any additional complicated steps, such as the typical annealing step for ITO films, and has yielded electrical and optical transmittance on par with standard TCO materials such as ITO and AZO. Furthermore, the materials used in this study amounted to about only half the indium than typical ITO films.

In comparison to other TCOs, the transmission is similar but some of the samples exhibited transmission in the IR region that is greater than reported for ITO and AZO, which is beneficial for PV applications. Optical transmittance in the visible region is greater than 80% with several samples demonstrating good transmittance in the near-infrared region with band-gaps ranging from 2.7 eV to 3.2 eV. The presented study shows that InZnO is capable as a transparent conducting oxide for PV applications. Future work for this study will be to fabricate devices with CIGS in comparison to other TCO materials and use an alternative CdS buffer.

References

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