

CHAPTER 6

INDIUM TIN OXIDE THIN FILMS: AN IDEAL TRANSPARENT AND CONDUCTIVE OXIDES CONTACT FOR SOLAR CELLS APPLICATION

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6.1 INTRODUCTION

Transparent and conducting oxides (TCOs) materials usually demonstrates simultaneously an exceptional properties such as high optical transmittance and good electrical conductivity[1]. TCOs materials have attracted a lot of interest, since after the first report of transparent and conductive cadmium oxide films in 1907 [2]. The increasing demands and advancement of optical and optoelectronic devices such as portable and flexible electronics, displays, solar cells, multi-functional windows and not long ago the transistors are some of the reasons for the huge interest [2,3]. Furthermore, the integration of different materials like metals, semiconductors, ceramics, plastics molecular and polymer organics into these devices have called for the provision of an improve TCO materials with required functional morphology, high performance and new processibility [5].

These materials are basically prepared from diverse type of materials including semiconducting oxides of cadmium, indium, tin, zinc and metals like gold, titanium nitride and silver [6]. Each of TCOs material reveals different properties depending on the constituent elements of the material. Examples of some commonly used TCOs are; fluorine tin-doped oxide (FTO), indium tin oxide (ITO), aluminium doped zinc oxide (AZO) and antimony doped tin oxide (ATO). One of the critical parameters that determine the choice of TCO is work function (Φ). It is the least energy required to remove an electron or other particles from the Femi level or the conduction band of material to the vacuum or into field-free space [7]. An excellent TCO utilized as front contact must have high work function required to serve as ohmic contact couple with high electrical conductivity, transparency and band edge position [8].

Indium tin oxide (ITO) is one of the leading TCOs material that has been studied extensively by researchers due to its versatile optical and electrical properties. Besides, ITO has a high chemical stability in many different environments and is widely considered for application in industries [6–10]. ITO ($\text{In}_2\text{O}_3:\text{SnO}_2$) is a direct and wide band gap ($\sim 4.5\text{--}5.1$ eV) material that exhibits high optical transmittance greater than 80 % in the visible region and low electrical resistivity to less than 10^{-4} $\Omega\text{-cm}$ [11–14]. In reference to these properties, for decades now, ITO thin films has been used as TCO contact in optoelectronics applications like solar cells[18], flat panel displays (FPD)[6], organic light emitting diodes (OLED) [16], liquid crystal displays, as antistatic material for cathode ray tubes (CRT) displays, electro chromic devices[21], ferroelectric photoconductor storage devices, thermocouple or thermoelectric element[22], infrared and radio frequency protective windows and heat reflecting mirrors[18, 20,21].

In the case of solar cells application, ITO films work

- (a) as anodes electrode used for extraction of separated charge carriers from the absorbing region of the semiconductor.
- (b) as a rectifying contact in which a simple silicon solar cell is formed or
- (c) as an antireflection coating for optimum transmittance of light [20].

It is also responsible for high absorption of energy in the absorbing part of solar cells, because of its high transmittance in the visible region. ITO can be easily patterned using well suitable techniques for device fabrications[24]. The stability property of ITO films is an advantage over other TCOs. Intrinsic oxides such as zinc oxide (ZnO), aluminum zinc oxide (AZO) and gallium zinc oxide (GZO) thin films have been shown to have relatively lower stability in term of electrical and chemical effects than non-doped ITO thin films [25]. Thus, TCO like AZO has some advantages for large-area application like low cost, non-toxicity, environmental stability and material abundance. But yet obtaining a lower resistivity in this material requires a high annealing temperature than in ITO[25,26].

6.2 PHYSICAL PROPERTIES OF ITO

ITO material have a cubic Bixbyite structure and the structure is formed when In^{3+} atoms are replaced with Sn, in which each tin and indium cation rests at the centre of the cube while the oxygen anions occupies the six corners of the structure as depicted in Fig. 6.1 [28]. The two oxygen vacancies greatly influence the ITO structural properties. High charge carrier concentration in ITO films is responsible for low resistivity since Fermi level (E_F) is above the conduction level (E_c) [29]. An electron is denoted by Sn atom to the carrier concentration thereby increasing the electrical conductivity of ITO. Similarly, The wide band gap energy of $\text{In}_2\text{O}_3:\text{SnO}_2$ is

responsible for it is high optical transmittance in the visible region whereas the good electrical conductivity is due to the arising intrinsic oxygen vacancies and extrinsic tin doping [1,30,31].

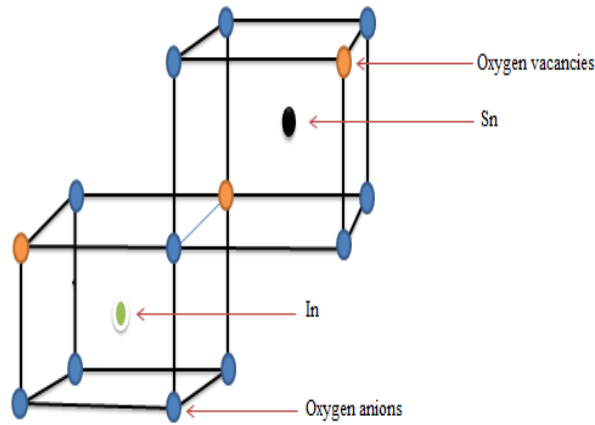


Fig. 6.1 A body centre Cubic Bixbyite structure of ITO [32].

ITO is a highly degenerate n-type semiconductor [33]. As a highly degenerate semiconductor, the large carrier concentration in ITO is due to its massive doping [34]. If the concentration of these carriers increases, impurity atoms distances decreases and it will reached a point when the donor impurities will begin to interact with each other[35]. Further increase in donor concentration will result in band widening of the donor states and Femi level E_F can now occupy the bottom of the conduction band as shown in Fig. 6.2 [36]. This E_F overlap usually occurs, when the donor charge concentration becomes comparable with the resulting density of electronic states. The electronics states close to conduction band minimum will now be completely filled as the E_F shifts to the higher energy levels within the conduction band. Hence, excitation of electrons from the valence band to available electronic states in the conduction band requires higher energy. This indicates that the increased in ITO band gap is a result of increased in carrier concentration [26,30].



Fig. 6.2 Schematic n-doped degenerate semiconductor energy-band gap with Femi level lie in the conduction bank.

Injection of holes into organic materials is done by high work function electrodes [37]. ITO work function in most of the optoelectronic applications plays an enormous roles in device performance as it affects the energy barrier height at the hetero-junction interface [28,38]. Lower potential energy barrier for hole injection is obtained when the work function of the anodes is high [37,39]. Hence, higher ITO work function is of very important since the lower barrier, brought about a sufficient efficiency in the injection of holes across the interface for improved device performance [27,37,40]. In a perfect anode material, the transmittance in the visible range and electrical conductivity are very high, surface contact is good, the work function is also high (>5 eV) [41]. ITO is usually used as high work function anode due to its excellent transparency and low resistivity for controlling the energy barrier for hole injection on interfaces between the ITO electrode and that of photoelectric or display organic material [38,40,42,43]. This process significantly affects the luminescence efficiency of the device [37,43].

6.3 DEPOSITION OF ITO THIN FILMS

Thin films deposition techniques play a huge role in fabricating films of good optical and electrical properties. Several techniques ranging from physical vapor deposition (PVD) and wet chemical deposition (WCD) methods were reported. Among the existence PVD method, magnetron sputtering, thermal evaporation and pulsed laser ablation are the most popular techniques used for ITO films deposition. These techniques rely on high vacuum systems ($> 10^{-7}$ Torr) in order to minimize impurities and contaminations. The type of the deposited films produced using PVD largely depends on the parameters used that characterize the deposition process which includes; vacuum chamber pressure and cleanliness, deposition rate, deposition atmosphere, and substrate temperature, target-substrate distance, substrate temperature and substrate angle [44].

Similarly, chemical deposition methods such chemical vapor deposition (CVD), sol-gel and spray pyrolysis methods have been applied in the depositions of ITO films. Most of chemical methods produced an amorphous ITO films of poor optical and electrical properties when operated at low substrate temperature of 150°C or less [45]. Sol-gel and spray pyrolysis methods are known as the more straightforward and cost-effective techniques, with more encouraging features of the latter like easy doping of several materials, high growth rate, and mass production ability for the vast uniform area. Of all these, the production of high quality and continuous films over a large area by magnetron sputtering technique have limited the use of this method [46–48].

Magnetron sputtering is a leading PVD technique and is widely considered for ITO films deposition [44]. This technique have prepared high-quality films with low resistance and high transparency for application in advanced optoelectronics devices. Magnetron sputtering has either direct current (DC) source or radio frequency (RF), both of which can get uniform film easily, with a good adherence to the substrate and low residual tensions, and is widely used in ITO film deposition [49]. It is a highly rated vacuum coating technique used for deposition of metals, alloys, or metal oxide compounds onto any types of materials with thicknesses up to about 5 μm [50]. This method is a low-pressure process, that has an advantage of fabricating uniform ITO films reproducibly with high electrical conductivity and visible transmittance [51, 52]. But blackening in ITO films is sometimes observed during the deposition of ITO films, which is attributed to the presence of indium metal [53].

In magnetron sputtering, the sputtering material is ejected due to the bombardment of ions to the target surface as shown in Fig. 6.3 [54]. In preparation of thin film deposition, argon ions are activated to sputter particles onto a substrate [55,56]. An inert gas such as argon filled off the vacuum chamber of the coating device, by which a glow discharge is activated when a high potential difference is applied, resulting in acceleration of ions to the target surface and a plasma coating [57]. Sputtering materials are ejected by argon-ion from the target material thereby resulting in a sputtered coating films layer on the substrate in front of the target [57,58]. Particles on the substrate gain additional energy, due to the conversion of high potential energy into kinetic energy, thereby facilitating crystallite nucleation, which can improve the optical and electrical properties of thin films [55,56,59].

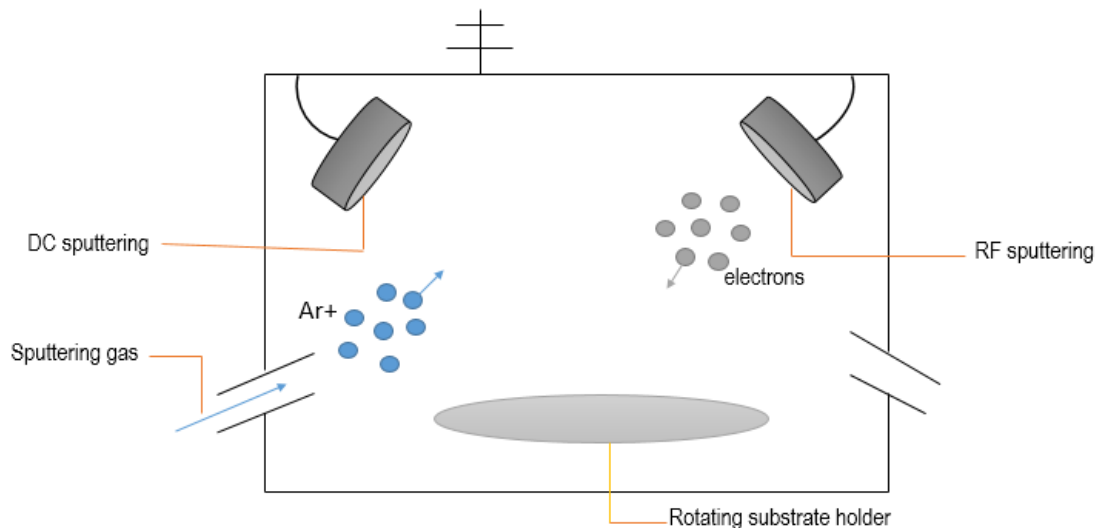


Fig. 6.3 Schematic diagram of dual magnetron sputtering system.

Usually, deposition of ITO films is carried out at temperature $> 250\text{ }^{\circ}\text{C}$, followed by temperature annealing at $> 300\text{ }^{\circ}\text{C}$ in order to improve the optoelectronic

properties of the contact [39]. Moreover, sputtered ITO films properties in the sputtering atmosphere are well sensitive to sputtering parameters like oxygen or hydrogen partial pressure, final sputtering pressure, substrate temperature and the distance between the target and substrate [60]–[63]. Kurdesau et al. [64] carried out a room temperature comparable study of RF and DC magnetron sputtering techniques with different optimization deposition conditions. At first, both the DC and RF sputtering produce high transparency in the visible range and low sheet resistance (20–25 Ω/sq) at room temperature. Different structural properties were obtained at optimized deposition conditions; with RF technique ITO layers showing strong crystalline X-ray diffraction peak intensities at improving conditions of higher discharge power density, Ar-O₂ gas mixture, and lower deposition rate. DC sputtering produces an amorphous ITO layers with nano-grain scale surface at optimized conditions of lower power density, high deposition rate and is carried out in pure Ar gas without any reactive gas addition.

6.4 ITO REQUIRMENTS FOR APPLICATION IN SOLAR CELL DEVICE

The incorporation of TCO thin layers in silicon (Si) solar cells is one of the progress so far achieved in the development of PV devices. These layers serve as an anti-reflection coating (ARC) if proper optical thickness can be obtained, junction rectifiers and transparent electrode for the extraction of separated charge carriers from the absorbing region of semiconductor material [1,65,66]. Recently, the TCO layer has also been deposited at the back of solar cell as backside reflector and it requires properties different from those use as front windows contacts [67]. The two most important properties of ITO films as contact in solar cell application are the optical transparency and electrical conductivity in which they are interrelated. It is observed that, when one property value is improved the other one value tend to decrease, hence there is a need for compromise [67]. Substrate heating during ITO films deposition or post-annealing treatments was reported to have improved the optoelectronic properties of the films thereby resolving the trade-off between transmittance and conductivity issue [56,68–70].

For Solar cell devices, ITO maximum transmittance is a crucial factor while reflectance should be as minimum as possible [32]. Several parameters of ITO thin film layer are considered in solar cells, such as electrical conductivity, optical transparency, thickness of the film, deposition rate and annealing temperature to reduce the sheet resistance and optical losses [17,18]. Moreover, good electrical conductivity and high optical transmissivity (450–650 nm wavelength) in the visible region couple with better quality ITO/Si interface are key requirements of ITO films in solar cell applications [71,72].

It is known that, one of the vital uses of ITO as TCOs in PV solar cell is a current collector (front window) in which its performance categorically depends on the type of solar cell. ITO and doped ZnO are the two most utilized TCOs in microcrystalline and amorphous silicon solar cells. Solar cells such as CIGS, Heterojunction with Intrinsic Thin layer (HIT), and dye-sensitized solar cell also utilizes ITO layer [32,73]. Additionally, the ability of ITO to forming a certain desired interface with Si material is another requirement. Since a simple efficient solar cell is obtained when rectifying contact is formed between ITO and Si [71][12].

6.5 TRANSPARENCY AND CONDUCTIVITY TRADE-OFF OF ITO FILMS

Transparent conductive contact (TCC) of good electrical conductivity and optical transparency are crucial components for various optical and electronic devices such as OLEDs and solar cells. For Solar cell devices, ITO maximum transmittance is a critical factor while reflectance is required to be very minimum. Several parameters are important in choosing a TCC thin layer for solar cells application, such as electrical conductivity, optical transparency, film thickness, deposition rate and temperature annealing. These properties generally influences films crystallizations which subsequently reduces films sheet resistance and optical losses [17,18].

ITO optical transparency and electrical conductivity are closely related such that increasing the former led to decreased in the latter. Absorption in the visible region of spectrum decreases with decreasing TCO thin layer which also lower the carrier concentration, and at the same, increases the sheet resistance R_{st} [74]. Realizing a low resistivity TCC requires the carrier mobilities and carrier concentrations to be as high as possible [75]. Hence, an increase in carrier concentration, and Hall mobility decreases the resistivity of TCC [76]. It is reported that the crystallinity and grain size of ITO films increases when ITO films were deposited at high substrate temperatures[77]. Increment in carrier concentrations (N) results in ITO band gap (E_g) widening, indicating the occupation of lowest states of the conduction band by excess charge carrier as explained by Burstein-Moss shift in Eq. 6.1 [24,78,79]. The energy band gas is given by,

$$E_g \propto N^{2/3} \quad (6.1)$$

In photovoltaic solar cells which is a semiconductor diode, band gap energy E_g which determines the amount of photon energy absorbed is a decisive parameter [73]. Transmissivity nature of any TCC is related to the wide band gap of that material [32]. High optical transmittance is required by any good TCC for free passage of light without much absorption [18]. For any TCC, two parameters are crucial: the sheet

resistance (R_{st}) and the optical transmission (Top), both of which depend on the coating thickness [26]. The sheet resistance is defined by,

$$R_{st} = 1 / \sigma t \quad (6.2)$$

Where σ is the electrical conductivity ($\Omega \text{ cm}$)⁻¹, and t is the coating thicknesses in cm. To evaluate the quality and performance of transparent conductive films with different thickness, resistivity, and transparency, a model called a figure of merit is used. Haacke,(1976) proposes a new figure of merit (FOM) in Eq. 6.4, after Fraser and Cook, (1972) had earlier proposed a FOM (Eq. 6.3) that results in maximum FOM values at large film thickness and low transmittance values which favors the sheet resistance [18]. FOM as proposed by Fraser and Cook is written as,

$$FOM = T / R_{st} \quad (6.3)$$

Where T is the optical transmission. However, the reversed version of FOM as developed by Haacke is,

$$FOM = T^{10} / R_{st} \quad (6.4)$$

From Eq. 6.4, it is observed that a higher value of FOM , results in the excellent performance of the film, which requires a low sheet resistance and high transmissivity [82].

6.6 ITO/ SILICON SOLAR CELLS

The application ITO layer on Si solar cell was successfully initiated in the late 1970s. Since then, many efforts have been made to improve the performance of the ITO/Si heterojunction (HJ) solar cells. ITO film in HJ solar cell serves as a front window for the separation of charge carriers and also as an antireflection coating if an appropriate thickness is found and recently, as backside reflector. Similarly, a simple efficient solar cell is achieved when rectifying contact is formed between ITO and Si [1,10,71]. As reported, there are good potential abilities in ITO/Si junction used for photovoltaic applications [32]. Largely ITO/p-Si or ITO/n-Si heterojunction was found to have a good response to short-wavelength photons compared to a diffused n^+p junction. This is possible since photons energy transmission through ITO covers both short and long wavelengths of energy that moves to photo-generation carrier junction. The highly degenerate n-ITO/c-Si junction known as degenerate semiconductor-insulator-semiconductor (SIS) cell has a good potential for PV applications [83].

The ITO/Si junction can either be a rectifying or an ohmic contact which solely depends on the method used for the deposition. For example, an ohmic contact is obtained when ITO film was deposited using spray pyrolysis on p-type Si and rectifying contact on n-type Si respectively[84]. When the ion-beam sputtering technique is used, an ohmic contact on n-type Si was achieved while on p-type Si, a rectifying barrier was obtained [85]. In general, high optical transmittance, low electrical resistivity, and high carrier mobility, good quality interface, non –toxic and good chemical stability are prerequisite to enhancing ITO/Si heterojunction. Fig. 6.4 shows incorporated ITO contacts in crystalline silicon solar cell.

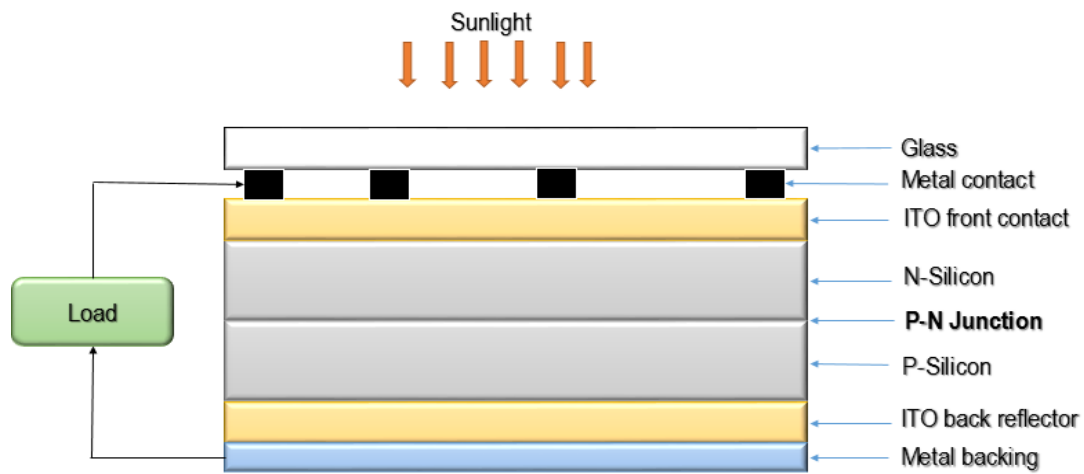


Fig. 6.4 Crystalline solar cell utilizing ITO as front window and back side reflector contacts.

Furthermore, for a good quality ITO/Si interface, free from generation/recombination centres, realisation of 20 % energy conversion efficiency is possible in ITO/Si heterojunction as estimated by Kobayashi et al. [86]. Ryu et al.[66], obtained an improved ITO/n+ emitter-Si contact quality with photo-conversion efficiency of 16.8 %. Prior to that, only a marginally above 10 % of conversion efficiency has been realised in ITO/Si junction [87][71]. The highest energy conversion efficiency of 26.3 % in a single crystalline silicon solar cells is achieved recently by Kaneka [88][89]. Moreover, the quality of the interface between ITO and Si junction is greatly affected by the existence of a thin interfacial layer in the junction. The presence of this interfacial oxide layer do not only initiates fixed charges but also increase the charges as the interfacial oxide layer widens[71]. This will categorically affect the ITO/Si junction electrical property. As stated by Kobayashi et al. [90] the ITO/p-Si photovoltaic effect increases with the existence of the thin silicon oxide layer (insulating layer).

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