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A Review of Metal Oxide Thin Films in Solar Cell Applications

Ho Soonmin[*]

INTI International University, Putra Nilai, 71800, Negeri Sembilan, Malaysia

Abstract: The metal oxide thin films have performed so greatly in semiconductor field because of excellent physical, optical and electric properties. These materials have attracted more and more interest, and could be used in solar cells, biosensors, biomedical, supercapacitor, photocatalysis, luminescent materials, and laser devices. Preparation of thin films have been reported by many researchers through various deposition methods. The characterization on the properties of films is very important to optimize its design of material. In this work, significantly focused on the development or improvement in thin film based solar cells, perovskite solar cells, and dye sensitized solar cell. The photovoltaic parameters (short-circuit current, open-circuit voltage, fill factor and efficiency) were studied in solar cell fabricated under different conditions. Experimental findings confirmed that metal oxide could be used as electron transport layer, electron conducting medium, anti-reflecting layer, and hole transport material.

Keywords: Thin film based solar cell, band gap, absorption, power conversion efficiency, semiconductor.

1 Introduction

In a nanotechnology field, the metal oxide semiconductors are used in biosensors [1], biomedical, supercapacitor [2], photo catalysis [3], luminescent materials [4], laser devices and solar cells [5-8]. Experimental results showed the properties of metal oxide thin films in optical, morphological, compositional, electrical properties and acoustical behaviors meet excellent applications in these fields. These films have been prepared on a variety of substrates via various deposition methods such as DC magnetron sputtering [9], chemical bath deposition [10], spray pyrolysis [11], sol gel spin coating [12], pulsed laser deposition [13], electro deposition [14], thermal evaporation deposition [15], molecular beam epitaxy [16], and chemical vapor deposition [17]. Researchers pointed out that each of these deposition techniques showed own advantages and limitations.

Solar energy is one of the renewable energy sources [18]. The solar energy is free and solar modules were operated without emission of any toxic gases [19]. Table 1 indicated first, second and third generation solar cells. Generally, working principle of these types of solar cells were based on the photo electric effect [20]. Currently, silicon remained the dominant photovoltaic material due to silicon

is non-toxic material, abundant and produce higher power conversion efficacy. However, the main drawback was too expensive. Later, development of thin film based solar cell was reported. Researcher highlighted that thin film technology can decrease the amount of active material in cell, and cheaper than crystalline silicon.

In this work, significantly focused on the development or improvement in thin film based solar cells, perovskite solar cells, and dye sensitized solar cell (DSSC). The performance of solar cell was investigated and photovoltaic parameters (open circuit voltage, short current density, fill factor and efficiency) were reported under various conditions.

2 Literature Survey

The zinc oxide (ZnO) showed n-type semiconductor materials and has wide band gap. ZnO films indicated easy crystallization, high carrier mobility, and technological flexibility. The solar cell was successfully made from carbon fiber/ZnO/epoxy resin/CuO/epoxy resin/carbon fiber [29]. The photovoltaic parameters such as open circuit voltage (Voc) (404 to 674.6 mV), short circuit current density (Jsc) (20.69 to 32.4 mA/cm^2), fill factor (52.33 to 62.12), and efficiency (5.88% to 13.57%) in different samples (under various composite film thickness, percentage of ZnO and

[*]Corresponding author E-mail: soonmin.ho@newinti.edu.my

PVA) were reported. The lower performance of solar cell was observed due to current carrier movement. The electron gained energy from photo electrode when the sunlight strikes light absorbing material (ZnO/PVA composite). The solar cell was fabricated using zinc oxide films with lead sulfide counter electrode. The short circuit current density was increased with increasing the growth time, due to the increased of the surface density of quantum dots [30]. The Jsc, fill factor, Voc and efficiency values reached 12.6 mA/cm^2, 0.61, 676 mV, and 5.19%, respectively. There are several deposition techniques have been used to produce Cu$_2$O/ZnO heterojunction solar cell [Table 2]. Power conversion

efficiency was highlighted for different solar cells.
The polyaniline-zinc oxide nanocomposites were used as material in solar cell. Based on the photovoltaic behavior investigations, the fill factor (48.57 to 65.36%), efficiency (1.683% to 2.69%), short circuit current density (7.7 to 8.1 mA/cm^2) and open circuit voltage (0.45 to 0.51V) are reported in different samples [36]. Sudheer and co-workers [37] pointed out ZnO films could be employed as electron transport layer in perovskite solar cell because of highly smooth, compact and uniform morphology.

Table 1: The first, second and third generation solar cells.

Solar cells	Description
First generation solar cells	• The crystalline and poly-crystalline silicon solar cells are examples of the oldest commercially available solar cells technologies [21]. • These types of solar cells contributed about 90% world solar cell market. • Silicon showed indirect band gap semiconductor material with band gap of 1.12 eV [22].
Second generation solar cells	• Copper indium gallium diselenide, copper zinc tin sulphide, cadmium telluride thin films, and amorphous silicon have been synthesized via different deposition methods onto the glass and flexible substrate. • These thin film based solar cells have many advantages including less material needed, low production cost, and low temperature large area deposition [23]. • These types of solar cells required photoactive materials (film thickness<1 μm), exhibited direct band gap with high absorption coefficient to absorb incident sunlight [24].
Third generation solar cells	• In dye sensitized solar cells, the organic and inorganic dye molecules were employed as sensitizers. These solar cells could be produced on rigid and flexible substrates [25]. • In bulk heterojunction solar cells, the C60 and conjugated conductive polymers were used as acceptor and donor materials, respectively [26]. • In perovskite solar cells, the light harvesting active layer was made from hybrid organic inorganic lead or tin halide based materials. Perovskite has specific crystal structure, reached highly efficient in a very short time [27]. • In quantum dot solar cells, the inorganic quantum dots were used as sensitizers, because of tunable nature of the band gap value [28].

Rameshkumar and co-workers [38] reported that tin oxide (SnO_2) films are suitable for solar cell applications. These materials can absorb maximum radiation in ultra-violet (UV), and then drops in visible region. The optical transmittance showed maximum transmittance about 70-80%, cut-off frequency is estimated to be 287 nm. The glass/FTO/SnO_2/TiO_2/P3HT:PCBM/MoO_3/Ag layer solar cell was designed [39]. The highest transmittance about 78%, can create electron hole pairs in the photoactive layers. Experimental results also revealed that these films are highly transparent, low surface roughness and excellent homogeneity. The photovoltaic performance including Jsc (10.4 mA/cm^2), Voc (0.78V), fill factor (35) and efficiency (2.87%) were highlighted in experiment. The SnO_2 microsphere with 75 nm diameter has been prepared using solvothermal route [40], indicated the highest light absorbing capability. The power conversion efficiency about 16.85%, with fill factor of 66.4%, Voc of 1.12 V and Jsc of 21.8 mA/cm^2. The smallest specific surface produced the lowest photovoltaic performance with increasing the microsphere size from 75 nm to 200 nm. Wang and co-workers [41] reported the SnO_2 microspheres (2.2 μm diameter) could be used as photo electrode for dye sensitized solar cells. Experimental findings supported that the best power conversion efficiency about 6.25%, fill factor=0.55, Voc=803 mV and Jsc=14.11 mA/cm^2. Govindhasamy and co-workers [42] concluded that SnO_2 films could be employed as electron conducting medium because of many reasons such as higher electron mobility, better anti-reflective behavior, wider band gap and more suitable band edges. The solar cell made from $CH_3NH_3PbI_3$ perovskite infiltrated SnO_2 indicated power conversion efficiency reached 8.38%, with fill factor of 45%, Voc of 0.96 mV, and Jsc of 18.99 mA/cm^2. Xiong and co-workers [43] described that SnO_2 could be used in perovskite solar cell because of deep conduction band and valence band. The power conversion efficacy successfully achieved 17% by using a low temperature sol derived SnO_2 films. However, they also explained that the limitations of SnO_2 as electron transport layer were easy degradation in high temperature and much lower conduction band may cause voltage loss in solar cell. Jung and co-workers [44] have reported hysteresis free planar perovskite solar cell by using SnO_2 films as electron transporting layer. The solar cell consisted of $(HC(NH_2)_2PbI_3)_{0.875}(CsPbBr_3)_{0.125}$, was produced on 40 nm thick, 0.1 M, 250 °C annealed SnO_2 films, reached the power conversion efficiency about 19.17%, fill factor of 0.74, Voc of 1.144V and Jsc of 22.64 mA/cm^2.

The cadmium oxide (CdO) is semiconductor of II-VI group, and it could be used in solar cell applications. The obtained films are high transparency in the visible area, electron concentration more than 10^{19}/cm^3, narrow direct band gap from 2.2 to 2.5 eV, excellent conductivity at room temperature, higher mobility of 130 cm^2/Vs. The solar cell consisted of Al/p-Si/n-CdO/Al was fabricated. Murugasamy and co-workers [45] reported that CdO films prepared at 400 °C indicated uniform and well developed small grains (102 nm) distributed over the entire film surface, can develop solar cell with high efficiency. Optical transmission spectra showed the CdO films maximum transmission about 60% at 650 nm, and transmission dropped in the UV region smaller than 400 nm. Experimental findings concluded that light absorption is moved towards lower wavelength side, especially for the large carrier concentration and highly non-stoichiometric materials. The fill factor and efficiency values increased from 0.31 to 0.32, and 0.6 to 0.8%, then, reduced to 0.3 and 0.7% for the films prepared at 350, 400 and 450 °C, respectively. Gaurav and co-workers [46] highlighted that CdO:TiO_2 could be employed as anti-reflecting layer, lead to improve the efficiency of heterojunction solar cells. Further, they explained these layers indicated highest conductivity and the lowest resistivity. The efficiency (3.23%), open circuit voltage (0.26V) and short circuit current (7.6 mA) were reported. Fahrettin [47] studied the photovoltaic properties of Al/p-Si/n-CdO/Al solar cell. They observed that photocurrent was increased when the illumination intensity was increased, because of more produced photo carriers. The best values of Voc and Jsc were estimated to be 0.41 and 2.19 mA/cm^2, respectively under AM 1.5 illumination. Atomic force microscopy was used to study the topography of nano crystalline CdO films [48]. The high degree of roughness (58.54 nm), decreases light reflection, increased light absorption in the visible region. The fill factor (0.652) and efficiency (5.5%) of n-CdO/p-Si were measured. Hasan and co-workers [49] have reported that the efficiency of Si/CdO solar cell could be improved after adding impurities atoms. Experimental results confirmed that Jsc, Voc, fill factor and efficiency were increased from pure CdO (9 mA/cm^2, 0.25 V, 0.346 and 0.78%), CdO:Sn (12 mA/cm^2, 0.3 V, 0.4 and 1.44%), CdO:Sb (15 mA/cm^2, 0.35 V, 0.43 and 2.24%) to CdO:Se films (17 mA/cm^2, 0.4 V, 0.51 and 3.5%). Muneer and co-workers [50] described the preparation of cadmium oxide (CdO) films by using chemical technique and drop casting method. X-ray diffraction (XRD) technique confirmed these films were cubic structure, polycrystalline and no trace of the other material. Photovoltaic properties revealed that Voc, Jsc, fill factor and efficiency were 4.1V, 1.44 mA, 36.2% and 6.8%, respectively.

Several researchers explained that nickel oxide played an important role as hole transport material. Because of some unique characteristics such as low cost, small hysteresis, acceptable band gap, good chemical stability [51], excellent transmittance, strong photo stability, low electrode polarization, and lower processing temperature for solar cell production. Rui and co-workers [52] reported that nickel oxide films were prepared via sol gel method, could be used as hole transporting material, to reach stable and high performance p-i-n structured quasi two dimensional perovskite solar cells. The maximum efficiency about 14.85%, Jsc of 17.95 mA/cm^2, and Voc of 1.12 V. Monika and co-workers [53] highlighted that NiO films have wide

band gap (3.19 eV), and can help in better ion transfer. The solar cell was fabricated and Isc was studied under different times such as 24h (1.5 mA), 48 h (1.2 mA), 96 h (0.9 mA) and 144 hours (0.8 mA). The best performance of solar cell with fill factor (0.45) and efficiency (14.04%) was reported. Shuangshuang and co-workers [54] found that higher power conversion efficiency could be observed in solar cell with doping transport layer if compared to un-doped transport layer. Further, they explained that charge separation and hole mobility derived from the perovskite/hole transport layer interface were successfully improved. The solar cell was fabricated by Ag/ITO/NiO/n-Si/Al carrier selective contact as pointed out by Mudgal and co-workers [55]. The photovoltaic parameters including Voc (509, 573 mV), Jsc (34.2 and 36.5 mA/cm^2), fill factor (712, 71 %) and efficiency (12.4 and 14.9%) were studied without and with the SiO$_2$ interlayer. Fei and co-workers [56] concluded that production of solar cell using nickel oxide indicated low fill factor and unideal Voc because of high charge trap density of deposited materials. Heterojunction solar cell consisted of p-type nickel oxide and n-type silicon was investigated under various experimental conditions [57]. The Voc (572.8 mV), Jsc (21.4 mA/cm^2), fill factor (54.37) and efficiency (6.67%) were improved when the NiO thickness was increased up to 0.1 μm. On the other hand, Voc was increased with reducing the buffer layer band gap because of reduce in valence band offset. Lastly, they reported that Voc, fill factor and efficiency achieved 906.16 mV, 69.82% and 12.73% with increasing the buffer layer thickness (from 0.001 to 0.01 μm). Boyd and co-workers [58] highlighted that photovoltaic performance under different hole transport layers. The Voc, Jsc, fill factor and efficiency were 1.12 V, 20.42 mA/cm^2, 80.8 %, 18.55% respectively by using poly TPD. The solar cell achieved Voc of 1.11 V, Jsc of 20.53 mA/cm^2, fill factor of 74% and efficiency of 16.63% by adding poly (trial amine) (PTAA). Finally, Voc, Jsc, fill factor and efficiency were 0.89V, 20.72 mA/cm^2, 79.2% and 14.57% for the solar cell designed with nickel oxide films. Yawen and co-workers [59] reported the solar cell made from NiO and PTAA under various concentrations. The power conversion efficiency reached 14.5%, 16.7%, 15.5% and 14.8% for 0.3 mg/ml PTAA/NiO, 0.5 mg/ml PTAA/NiO, 0.7 mg/ml PTAA/NiO, 1 mg/ml PTAA/NiO, respectively.

Chao and co-workers [60] reported the magnesium oxide (MgO) layer can passivate surface defects and also modify the heterojunction interface. Research findings showed the photon generated electrons rapidly tunnel via the MgO layer, the back recombination was blocked. Photovoltaic behaviors of Cu$_2$O/SnO$_2$ solar cells under various MgO thickness were studied. The Jsc (2.01 to 3.2 mAcm^{-2}), Voc (0.28 to 0.44 V), fill factor (28.64 to 36.9 %) and efficiency (0.16 to 0.51%) were increased with increasing the MgO thickness from 0.5 nm to 1 nm. Experimental results confirmed that Jsc and fill factor reduced in thicker MgO layer (2 nm), because of larger series resistance. The photovoltaic parameters of the solar cell under different precursor concentrations were reported [61].

The efficiency (10.3 to 11%), fill factor (69.6 to 71.5%), Jsc (18.75 to 19.28 mA/cm^2) and Voc (0.79 to 0.8 V) increased when the precursor concentrations were increased from 0.01M to 0.03 M. Analysis showed the performance of solar cell degraded rapidly with increasing the concentration up to 0.05 M. Ashish and co-workers [62] highlighted the MgO incorporated into TiO$_2$ perovskite solar cell, improved Voc from 0.86 V to 0.98 V Experimental findings supported that recombination at FTO/TiO$_2$ interface is very important factor regulating the voltage of solar cells. The solar cell was made from ITO/SnO$_2$/MgO/CH$_3$NH$_3$PbI$_3$/Spiro-MeOTAD/Au [63]. The MgO was prepared by using magnesium acetate tetrahydrate in the presence of ethanol over the SnO$_2$ layer via spin coating method. The highest efficiency reached about 19% under a sun illumination.

Researcher concluded that the efficiency of the solar cell could be improved by adding dielectric coating (such as Al$_2$O$_3$) on the top of transparent conductive oxide (TCO) layer. The Al$_2$O$_3$ showed high dielectric strength, good stability, excellent transparency and toughness under harsh conditions [64]. The solar cell was made from Al$_2$O$_3$/ITO (double layered anti-reflection coating) showed enhancement in efficiency (from 20.95% to 21.6%), external quantum efficiency (76.89% to 84.34%), Jsc (from 39.9 to 41.13 mA/cm^2) and reduction of average reflectance (from 9.3% to 4.74%). The influence of double layer anti-reflection (DLAR) and triple layer anti-reflection (TLAR) coatings on the performance of solar cell was studied [65]. The TLAR coatings exhibited higher fill factor (88.98%), efficiency (32.71%), Jsc (12.15 mA/cm^2) and Voc (2.74) if compared to DLAR coatings (88.51%, 30.65%, 11.28 mA/cm^2 and 2.73V). Researcher highlighted that perovskite was used as light absorber and hole transporter and in solar cells. Naser and co-workers [66] reported that the Al$_2$O$_3$ were used as mesoporous scaffold for production of solar cell. Research findings showed the collection efficiency increased with increasing the average roughness, and reducing the scaffold thickness. Liang and co-workers [67] described insulating ultra-thin Al$_2$O$_3$ interfacial layer between TiO$_2$ films and perovskite layer. If the thin films were too thick, leads to increase the electron transfer barrier. The performance of solar cell was studied with varying Al$_2$O$_3$ soaking times. The Voc (0.86 to0.93 V) and Jsc (17.21 to 21.16 mA/cm^2) values increased with increasing soaking times (from 0 to 60 minutes). Power conversion efficiency reached 9.36%, 10.2%, 12.79% and 11.32% when the soaking time was 0, 10, 30 and 60 minutes. The new passivation stack based on Al$_2$O$_3$ and SiC:B was created by Bernd and co-workers [68]. In this experiments, Voc, Jsc, fill factor and efficiency values are in the range of 670 to 681.8 mV, 37.9 to 40.5 mA/cm2, 75.9 to 82.5%, and 20.5% to 21.4%, respectively. The Al$_2$O$_3$ films (as moisture barrier layer) were produced by using plasma enhanced atomic layer deposition. According to experimental results, non-encapsulated cells indicated low shunt resistance (experienced degradation) if compared to Al$_2$O$_3$-encapsulated cells (stable during the test). These results confirmed that 50 nm Al$_2$O$_3$ layer can protect GaInP/GaAs/ge-Al$_2$O$_3$ solar cell from moisture during 1000

hours experimental time at 85 °C and 85% RH [69]. Choi and co-workers [70] produced Al_2O_3 by using atomic layer deposition method which could be used as encapsulant for perovskite solar cells. Experimental results confirmed that PTAA based solar cell experienced less than 4% reduced in solar cell after 7500 hours of 50% RH at room temperature. Because of the presence of pinholes within spiro-OMeTAD layer after thermal stress. Tu and co-workers [71] reported that the reflectivity of Al_2O_3 and TiO_2 reduced but, reflectivity of MgF_2 increased with increasing of refractive index of the films. Also, they concluded that reflectivity of $TiO_2/Al_2O_3/MgF_2$ multilayer AR coating reduced in 750-900 nm wavelength, indicating degradation spectral window for triple junction solar cells.

catalytic activity while transition metal showed poor electron transport efficiency between particles. Ming and co-workers [72] reported the development of Co_3O_4 and Co_2N supported on mesoporous carbon, which produced via facile oxidation and nitridation treatment process. The Voc, Jsc, fill factor and efficiency values are in the range of 0.688 to 0.697 V, 14.01 to 16.26 mA/cm^2, 0.23 to 0.45 and 2.42 to 5.08 %, respectively in Co_3O_4 composites. On the other hand, the photovoltaic parameters such as Voc (0.707 to 0.724 V), Jsc (8.6 to 12.6 mA/cm^2), fill factor (0.51 to 0.58) and efficiency (3.08 to 5.26%) values were highlighted also in Co_2N composites. Ambika [73] described that $Co3O4$ could be used in solar cell due to

Table 2: Power conversion efficiency of ZnO/Cu_2O heterojunction solar cells produced by using various deposition techniques.

Deposition technique	Solar cell	Power conversion efficiency (%)	Reference
successive ionic layer adsorption and reaction	ITO/NiO/Cu$_2$O/ZnO/SnO$_2$/Al	1.12	Chatterjee and co-workers, 2016 [31]
electro deposition	FTO/ZnO/Cu$_2$O/Au	1.43	Fujimoto and co-workers, 2013 [32]
Thermal oxidation/PLD	Cu/Cu$_2$O/ZnO/AZO	3.85	Minami and co-workers, 2011 [33]
Galvanostatic electro deposition	FTO/ZnO/Cu$_2$O/Au	0.41	Jeong and co-workers, 2008 [34]
Magnetron sputtering	IGZO/ZnO/Cu$_2$O/Au	1.68	Ke and co-workers, 2017 [35]

well dispersed with high crystallinity. The performance of solar cell including Voc (0.812V), Jsc (19 mA/cm^2), fill factor (0.69) and efficiency (9.05%) were reported. The development of p-type inorganic materials such as cobalt oxide as hole transport layer (to replace organic hole transport materials) due to great stability, fast hole mobility (10^{-2} to 10^{-1} cm^2/V.s), superior capability in blocking water and oxygen. Aibin and co-workers [74] highlighted

The platinum can be used as counter electrode in the dye sensitized solar cell because of good conductivity and excellent catalytic activity. However, platinum showed some limitations such as high cost and limited resources. The carbon materials and transition metal can replace the platinum because of abundant and cheap materials. The disadvantage of carbon materials was low intrinsic electro-

ultrathin hole transport layer (CoO film thickness=10 nm) in work in order to minimize incident light loss caused by cobalt ion absorption, and decreased the carrier transport loss by shortening the transport path. The maximum power conversion efficiency of solar cell achieved 10%. The solar cell consisted of FTO/CoO/perovskite/PCBM/Ag was reported [75]. The solar cell based on the untreated CoO indicated poor performance (Voc=0.828V, Jsc=10.8 mA/cm^2, fill factor=24.9%, efficiency = 2.2%) if compared to treated CoO films (Voc=0.933V, Jsc=18.9 mA/cm^2, fill factor=57.2%, efficiency = 10.1%). Shalan and co-workers [76] concluded that CoO films are excellent hole extracting layer. The power conversion efficiency reached 14.5% from ITO/CoO/CH$_3$NH$_3$PbI$_3$/PCBM/Ag solar cell, higher than traditional hole extracting layer including NiO (10.2%),

PEDOT:Pss (12.2%), and CuO (9.4%). Fabrication of stable perovskite solar cell by using CoO nanoplates. Dou and co-workers [77] pointed out that uncovered perovskite crystalline grains can guarantee the unobstructed transport of holes from perovskite layers to hole transport layers. The performance parameters of the solar cell based on different counter electrodes were studied [78]. The Voc, Jsc, fill factor and efficiency were 0.655 V, 20.42 mA/cm^2, 0.45 and 6.02% and 0.598V, 17.75 mA/cm^2, 0.38 and 4.06% in CoO electrode and Cu$_2$S elecrode, respectively. Enbo and co-workers [79] described Co$_3$O$_4$-WC-CN/rGO which can be used for the counter electrode in the solar cell. The CN is nitrogen doped carbon, and rGO is reduced graphene oxide. The power conversion efficiency reached 4.53%, 6.32%, 5.6% and 7.38% when Co$_3$O$_4$-CN, Co$_3$O$_4$-WC-CN, Co$_3$O$_4$-CN/rGO, and Co$_3$O$_4$-WC-CN/rGO were used as counter electrodes, respectively.

The tungsten oxide (WO$_3$) was employed as hole extraction and transport layer in various types of solar cells such as organic, polymer and bulk hetero junction solar cells. Solution processed tungsten oxide films have been employed as active anode buffer layer in polymer solar cells. Dip and co-workers [80] reported that tungsten oxide films have high band gap (3.5 eV) and could be classified as direct band gap materials, which could be used for window layer in solar cell.

3 Conclusions

The metal oxide thin films have been attracted much attention in solar cell applications. Several deposition methods have been used to produce these films. The characterization on the properties of films is very important to optimize its design of material. This review has summarized photovoltaic parameters about various types of solar cells prepared under different experimental conditions. The obtained results highlighted metal oxide could be used as electron transport layer, electron conducting medium, anti-reflecting layer, and hole transport material in solar cell application.

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