

### Review Article

## The Role of Physical Techniques on the Preparation of Photoanodes for Dye Sensitized Solar Cells

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Dye sensitized solar cells (DSSCs) have attracted numerous research, especially in the context of enhancing their efficiency and durability, due to the low-cost and environmentally friendly nature of photovoltaic (PV) technology. The materials in DSSCs are vital towards the realization of these goals, since many of the important components are influenced by their respective preparation and deposition methods. This review aims to detail the research and development aspects of the different physical methods with the purpose of evaluating their prospects and corresponding limitations. The diversity of consideration and criteria includes thin film applications, material characteristics, and process technology that need to be taken into account when selecting a specific deposition method. Choosing a deposition method is not as simple as it seems and is rendered quite complicated due to various factors. Usually, a researcher will evaluate techniques based on factors such as the different preparations and deposition technology with materials' and substrates' type, specified applications, costs, and efficiencies.

#### **1. Introduction**

Dye sensitized solar cells (DSSCs) in the form of novel photovoltaic (PV) technology have widely huge potential for low-cost and environmentally friendly solar cells compared to their more traditional counterparts. DSSCs are favored, due to properties such as low weight, flexibility, transparency, varied colors, and superior performance in darker conditions, and have attracted considerable private sector investment and government funding. DSSC is considered as a promising research topic, due to the dependence of its power conversion efficiency on its materials. However, the materials [1] alone are not enough to guarantee a stable, efficient, and low-cost dye sensitized solar cell [2–4], as its properties are intertwined with the preparation and deposition method. It has been prepared via various deposition technologies, and these technologies are being intensely researched and

developed, due to their unique advantages over others. The purpose of this review highlights the different preparations and deposition methods that have been utilized in DSSC applications in order to emphasize their advantages and disadvantages, which will allow a researcher to select and optimize a given method. Moreover, other parameters such as sensitization and curing procedure play a significant role in DSSC's performance, which must be taken into account as well [5, 6]. There are two major categories in the preparation and deposition methods, including chemical and physical methods. We analyzed the chemical methods in our previous paper [7], and this review will attempt to discuss the physical applied methods in the preparation of DSSC. Meanwhile, the combination of both physical and chemical reactions is being considered in the preparation of DSSCs; however, this review is limited to physical preparations.

#### 2. Physical Methods

Physical methods utilize physical phenomenon to prepare and deposit the materials. Physical methods are further categorized according to their medium or precursors, such as gas or liquid. We tried to categorize these methods by considering this concept, although it does overlap from time to time.

#### 2.1. Liquid Phase Precursors

2.1.1. Spin Coating. Uniform thin films on flat substrates are deposited via the spin coating method, which encompasses products such as thin and ultrathin polymer films. Procedures on spin coating include deposition, spinup, spinoff, and evaporation (Figure 1). The overloaded solution is deposited onto the substrate, rotated at high speeds, and finally, the solution coats the substrate via centrifugal force. The desired thickness is obtained by continuous rotation. The solvent, being volatile, simultaneously evaporates. The film's thickness is dependent on the solution's concentration, solvent, and spin speeds [8]. The thin film is deposited by the high angular spin speed. The thickness of the films is less than 10 nm, which is useful in microfabrication and photolithography, which requires the thickness of the photoresist to be circa 1 micrometer. The film thickness is explained as follows [9]:

$$h = \left(1 - \frac{\rho_A}{\rho_{A0}}\right) \cdot \left(\frac{3\eta \cdot m}{2\rho_{A0}\omega^2}\right)^{(1/3)},\tag{1}$$

where *h* is thickness,  $\rho_A$  is density of volatile liquid,  $\eta$  is solution viscosity, *m* is evaporation rate, and  $\omega$  is angular speed. For the experimental results,  $h = A\omega^{-B}$  can be used, where *A* and *B* are constant parameters and B = 0.4 and 0.7 in most cases. The desired thickness, uniform film, simple setup, and low power consumption are some of the advantages of this method. However, its dependence of the substrate's size of the spin coating process, the lack of material sufficiency, high cost, and its requirement for a flat substrate are some of its known disadvantages [10]. As can be seen in Table 1, the list of materials could be used via the spin coating method in DSSC application.

2.1.2. Dip Coating. Dip coating results in a uniform thin film of liquid deposition upon the substrate for small slabs and cylinders (Figure 2). The thickness of the film depends on the viscous force, capillary (surface tension) force, and gravity. The flow conditions of the liquid bath and gas overhead play an important role in both thicknesses and uniformities. Thicker films resulted from using volatile solutes and combining rapid sufficient drying via basic liquid flow. The growth, theoretical prediction, and control of the process represent some of the challenges faced when utilizing this method. If the withdrawal speed is chosen in a way that the sheer rates remain with the Newtonian regime, the thickness deposition



can be calculated by the Landau-Levich equation, as follows [14]:

$$h = 0.94 \cdot \frac{(\eta \cdot \nu)^{2/3}}{\gamma_{\rm IV}^{1/6} (\rho \cdot g)^{1/2}},\tag{2}$$

where *h* is coating thickness,  $\eta$  is viscosity, *v* is dragging speed,  $\gamma_{\text{LV}}$  is liquid-vapor surface tension,  $\rho$  is density, and *g* is the gravity constant.

The dip coating process is divided into five steps [15]. (1) Immersion: the substrate is immersed in the coating material solution at a consistent speed for a moment, and (2) startup: the substrate is subsequently pulled up, which precipitates the formation of a thin layer to form on the substrate. Pullingup is performed at a steady speed to eliminate any potential jitters. (3) Deposition: the thickness of the coating is controlled by the pull speed, as a thicker coating will form if the pull (withdrawal) speed is increased. If a thinner layer is required, then (4) drainage: the excess liquid is drained, and (5) evaporation: any remaining solution on the coating is evaporated using volatile solvents such as alcohols. This step is continuously repeated. The dip coating method is rather common, while the spin coating method that was previously discussed is more suitable for industrial purposes [16]. The dip coating method is used in DSSC applications, such as Poly(N-vinyl-2-pyrrolidone)-capped platinum nanoclusters deposited on indium tin oxide glass (efficiency, 2.84%) [17] and mesoporous TiO<sub>2</sub> films deposited on an F-doped tin oxide with dye (ruthenium complex bis-tetrabutylammoniumcis-dithiocyanato-N,N'-bis-2,2'-bipyridine-4-carboxylic acid, 4'-carboxylate ruthenium(II)) (efficiency, 10.80%) [18].

2.1.3. Doctor Blade Printing (Knife to Edge) Roll to Roll. Ceramic capacitors are the first recorded tape-casting machines that were made by Glen Howatt [19], and this marvelous invention is soon followed by the doctor blade or tape casting method. The multilayer ceramic capacitors (MLCCs), low temperature cofired ceramics (LTCCs), lithium batteries, fuel cells, oxygen separators, and thermistors are some of the devices that are manufactured by the tape casting method. The application of this method vis-à-vis thin films is relatively recent, which found its niche in nanotechnological advances

Mataziala	Efficiency	Deference
Materials	η (%)	Reference
Poly-3-phenylhydrazone thiophene (PPHT):ZnO/PPHT:Dye:ZnO	0.55	[64]
PEDOT:PSS film on FTO glass substrate	6.37	[65]
ZnO nanowires	5.20	[66]
Pt electrodes	7.18	[67]
7,14-Bis[2-[tris(1-methylethyl)silyl]ethynyl]dibenzo[ <i>b,def</i> ] chrysene (TIPS-DBC) layers on SWNT/FTO	_	[68]
MWCNT on FTO	4.94	[69]
TiO <sub>2</sub> nanoparticles/nanofibers bilayer film	4.75	[70]
Nanocarbon composite with hierarchical porous structure	6.50	[71]
Camphorsulfonic acid doped polyaniline (PANI/CSA) film	2.67	[72]
Mesoporous TiO <sub>2</sub>	6.92	[73]
Al <sub>2</sub> O <sub>3</sub> and SiO <sub>2</sub> nanoparticles on ZnO nanowire and nanoparticles	2.40	[74, 75]
PEDOT:PSS (transparent thin films)	_	[76]
TiO <sub>2</sub> /NiO composite particles	3.27	[77]
Thin film of ZnO on FTO (2Ml)	6.70	[78]
ZnO compact film	7.76	[79]
TiO <sub>2</sub> film	_	[80]
A polystyrene (PS) film of microporous or island-like structure	4.58	[81]
Graphene/PEDOT-PSS	4.50	[82]
Thin films of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS)	0.34	[83]
Carbon-modified TiO <sub>2</sub> electrode	5.60	[84]
TiO <sub>2</sub> /PEDOT	0.93	[85]
SnO <sub>2</sub> colloidal suspension (18 nm)	_	[86]
Oleic acid graphite (OLA-G) films on FTO glass	0.77	[87]
Spiro-OMeTAD (2,2',7,7'-tetrakis-(N,N-di-p-methoxyphenylamine)-9,9'-spirobifluorene)	3.02	[88]

TABLE 1: List of materials was prepared via spin coating method for fabrication DSSC using some research results.



FIGURE 2: Scheme of dip coating process: (a) immersion, (b) wetting, and (c) withdrawal [12].



FIGURE 3: (a) A photograph of doctor blading (ErichsenCoatmaster 509 MC-I) [11] and (b) First step: the colloidal solution is deposited onto glass substrate; second step: the blade spreads the solution over the substrate; third step: the obtained film is dried [13].

in ceramic body armor, optical films, medical films used as drug delivery systems, and microporous membranes.

The well-defined thickness of films fabricated using the doctor blade technique is detailed in Figure 3. This method, aptly named tape casting, knife coating, or knife-over-edge coating, due to transfer to reel-to reel coating (R2R), is also viable to be used to make films for polymer solar cells [20]. The stages of this technique are summarized as the movement of a substrate at a constant speed under a blade, with a specific height and contact angle. The function of this blade is to spread the paste onto the surface homogeneously and get a fixed thick film. We can also conduct the process by placing a fixed amount of paste over the substrate to be deposited and then unrollis, using a glass rod, is moved on the spacer on both sides to determine the thickness of the film.

This method is economical, and it minimizes the loss of particles by around 5% compared to its spin coating counterpart. As a result of this, only a small amount of starting materials is sufficient for this method [11]. The coated thickness is around  $10-500 \,\mu$ m using a sharp blade at a fixed distance from the substrate's surface. The wet thickness of the film depends on the surface tension and the viscosity of the coating solution. In practical terms, this technique is analogous to spin coating, due to the enhanced initial loss of coating solution, and the extended time that is required to determine the best coating parameters. However, it is considered as one of the most straightforward and low-cost process for semi-conductor paste deposition and one of the simplest printing methods available for modern electronics.

This method is suitable for mass production of electroceramic thick films, as it requires less starting materials compared to the spin coating method. Despite its benefits, there are some areas where it is lacking, such as its slow evaporation or its tendency to aggregate or crystallize at high solution/paste concentration. The large effective area of the photovoltaic devices is based upon the compatibility between the accepters and donors. For example, the best combination between fullerene as an acceptor and polymer (paraphenylenevinylene, PPV) as a donor material is viable for the doctor blade method without any loss of efficiency [21]. This process could be performed manually in the laboratory (Table 2). The researchers have built simple and quick samples of nanostructure semiconductor films, and this method also allows science hobbyist to construct homemade solar cells.

2.1.4. Screen Printing. The screen printing process is twodimensional print layer patterning devised in the 20th century, which was determined to have numerous industrial applications. First, the porous printing plate is fabricated using a woven mesh of synthetic fabric threads or metal wire, which is duly mixed with stencil as a masking material. The process is detailed in Figure 4. The woven mesh is attached to a rigid aluminum or steel framework, which has a rectangular plane in a majority of the applications. However, the cylinder screen is also considered to be attached and sealed at both ends. The semirigid planar surface creates tension in the opposite direction of the mesh, similar to

TABLE 2: The summarized materials are used by doctor balding method for DSSC application.

Materials	Efficiency η (%)	Reference
Blends of polymer and fullerene		[21]
Porous carbon/SnO <sub>2</sub> /TiO <sub>2</sub> nanocomposite films	6.15	[89]
Sn doped ZnO nanostructures	1.82	[90]
ZnO photoelectrodes	1.07	[91]
ITO-PEN	4.24	[92]
ZnO porous film	4.64	[93]
TiO <sub>2</sub> layer	8.10	[94]
Carbon counter electrode (CE)	6.46	[95]
Porous carbon counter electrodes	6.10	[96]
TiO <sub>2</sub> film was coated on a Ti-isopropoxide-treated stainless steel-based substrate	8.30	[97]
Cauliflower-like TiO <sub>2</sub> rough spheres	7.36	[98]
Porous ZnO photoelectrode	4.50	[99]
Mesoporous TiO <sub>2</sub> film	5.80	[100]
FTO glass	9.10	[25]
ZnO and TiO <sub>2</sub>	1.60	[101]
Mesoporous TiO <sub>2</sub> electrodes	0.50	[102]
Pure anatase TiO <sub>2</sub> nanocrystallites	5.65	[103]
TiO <sub>2</sub> porous film	3.10 (glass substrate) 4.90 (plastic substrate)	[104]
TiO <sub>2</sub> on FTO glass	5.18	[105]
TiO <sub>2</sub> photoanodes FTO glass	7.21 (front side) 4.85 (back side)	[106]

the tensions generated via the interaction between synthetic polyester monofilaments or stainless steel. This stretched printing screen achieves three discrete purposes: (1) the fluid coating meters under pressure, (2) providing a surface for the shearing of the coating material viscous columns to transfer to the substrate, and (3) providing support for the imaging elements (the stencil). This method eliminates any loss of the coating solution and produces a large wet thick film with a high viscosity.

The screen printing method uses thick film printing, producing films that are around 10–15 microns. Example of application of this method includes the DSSC electrode, with TiO<sub>2</sub> as thick film deposited on a conductive glass (FTO/ITO). This method is widespread in the electronic industries to manufacturing DSSC in producing miniature and robust electronic circuits in a cost-effective manner, massive and automated, well-defined, and with highly reproducible structures. First, the thick film screen printing ink or paste is pressed onto the substrate by a mechanical squeegee, transferring the patterns [22]. This method consists of a solvent (organic or inorganic salt) and a binder (TiO<sub>2</sub> particles). The role of the solvent provides an ink homogeneous mixture for printing purposes. The screen includes woven mesh of

stainless steel, nylon, or polyester mounted under tension on a metal frame. During printing, the substrate is held at a distance from one side of the screen and the ink is deposited on the screen from the opposite side, while a squeegee traverses the screen under pressure. Second, the solvent will be dried at 120°C, which automatically guarantees the stability of the printed films and substrates. The nonvolatile portion of the solvent evaporates under high temperature. The most popular screen printable ink recipe contains ethanol for its solvent, terpineol for its dispersant, ethyl cellulose for its thickener, and Triton X-100 for its surfactant [23]. The water solvent causes cracks to form on the film, so organic solvents are used. The long stability and superior morphology of TiO<sub>2</sub> are important in dye adsorption, which utilize terpineol component and ethyl cellulose, respectively. The loading nanoparticles on screening print are important, because they affect the TiO<sub>2</sub> structure, although they reduce the efficiency of DSSC if the dispersion is incomplete. The low current output and the decline in the efficiency of the solar cell are due to the agglomeration of nanoparticles. The thickness of the coated film is superior compared to the other methods, such as offset lithography, gravure, flexography, xerography, or ink-jet printing, caused by the flow of coating material being under pressure into and through this mesh or matrix via coating on the substrate. The high efficiency for DSSC is around 9.1% for porous TiO<sub>2</sub> [24] and carbon black [25]. The ZnO nanoparticle shows an efficiency of 1.50% [26], whereas ZnO nanoparticles with two indoline dyes, coded D149 and D205, observe higher efficiencies at 4.95% (D149) and 5.34% (D205), respectively [27]. The ITO-PEN [28] and Pt deposited on ITO/polyethylene naphthalate (ITO/PEN) [29] have efficiencies of 5.41% and 7.20%, respectively.

2.1.5. Electrohydrodynamic Deposition (Electrospray Deposition; ESD). The liquid-phase process is a new method for fabricating thin films that are thinner than 10  $\mu$ m in MEMS (Microelectromechanical Systems) and NEMS (Nanoelectromechanical Systems) (Figure 5). The electrospray forms droplets from the sample solution, such as nanoparticles, and these droplets will be deposited on various substrates. The uniform solid film is formed by evaporating or heating the solvent on the surface via sintering. The thin films could be formed by a precursor compound, which decomposes at high temperatures or converts to another substance via chemical reactions with other compounds that are simultaneously sprayed or delivered during its gaseous phase [30]. This method could be used in physical and chemical reactions that are taken into account in the physical aspect in this review. The metal nitrates or acetates dissolve in water, methanol, and ethanol, and these mixtures are electrosprayed as precursors for a metal-oxide layer production. The advantage of this method is the usage of the less soluble materials [31], the ability to control charge droplet, the lack of solvent evaporation effects, and the ability to form multilayered thin films. The significant advantage of ESD is the multilayered thin film growth being superior compared to spin coating, dip coating, and screen printing, since the bottom organic layer solvent during the top layer growth cannot be avoided



FIGURE 4: Schematic of screen printing [11].



FIGURE 5: Schematic of electrospray deposition (a) from solution and (b) from precursor [30].

[32]. The nanocrystalline TiO<sub>2</sub> electrodes [33], porous TiO<sub>2</sub> films [34], and terpyridine-coordinated dye [33] had the efficiencies of 5.97%, 5.10%, and 2.48%, respectively. The spray dye molecules onto TiO<sub>2</sub> surfaces used this method to develop molecular dye sensitized solar cells [31]. High conductivity (electrical resistivity of  $2 \times 10^{-4} \Omega$  cm) and high transmission of 78% in the visible range were observed in Tin oxide films [35].

#### 3. Gas Phase Precursors

3.1. Physical Vapor Deposition (PVD). Thin film deposition via PVD is regarded as a vacuum coating technology and is classified into two techniques, evaporation and sputtering. The material could be coated in an energetic and entropic environment, since the particles will escape from the surface. The solid layer that is formed on the surface is due to the cooling of the particles once they lose their energy when immobilized. The particles are capable of moving in a straight path because the system is kept in vacuum, and the films are coated by physical means that are commonly directional, rather than conformal, in nature. Examples of PVD method prepared nanostructures in DSSCs are mesoporous TiO<sub>2</sub> nanocolumnar films [36] (3 mm-linear: efficiency (2.40%), 3 mm–500 nm: efficiency (2.20%), 3 mm–100 nm: efficiency (2.30%), 5 mm–500 nm: efficiency (2.50%), and 6  $\mu$ m reference P25 electrode: efficiency (2.10%)), ZnO nanostructures deposited on glass ITO substrate (efficiencies (0.10%–0.50%) for nanowires, nanowire sheets, branched Au, and branched Zn [37]), TiO<sub>2</sub> (nanotubes) deposition enhance dye loading to up to 47.20% and changing the efficiency to 6.28% for DSSC [38], and TiO<sub>2</sub> electrodes deposited at different angles (500 nm thickness: efficiency (0.60%–1.04%) with modified angle (60°–85°) and 3  $\mu$ m thickness: efficiency (2.78%) [39]).

*3.1.1. Evaporation.* Generally, thin films are deposited by evaporation. The source material is evaporated in a vacuum, which allows the vapor particles to travel directly to the substrate as a target and condense into a solid state.

*Thermal Evaporation.* Thermal evaporation is a simple and economical method in fabricating thick films and microfilms (Figure 6). The source materials are melted or sublimated via electrical heating, converting it into a gaseous phase.

TABLE 3: Thermal evaporation method used for DSSC application.

Materials	Efficiency η (%)	Reference
ZnO nanocombs onto FTO substrate	0.68	[107]
ZnO:Sb fiber on FTO	_	[108]
ZnO with different thickness of NiO thin films	1-0.9	[109]
Arrays of ZnO nanowires	0.34-0.85	[110]
Mesoporous TiO <sub>2</sub>	_	[111]
ZnO films onto FTO	1.56	[112]
ZnO nanofibers	0.88	[113]
Ag nanowires	2.70	[114]
Pt thin film on FTO	2.70	[115]



FIGURE 6: Schematic of thermal evaporation [40].

The vapor particles will then achieve a certain velocity in the chamber and will attempt to condense on the substrate. The substrates are located close to the source materials. The high vacuum condition in the vacuum chamber will act to prevent the particles from scattering and minimize the residual gas impurities. The temperature is kept between 1000~2000°C. There are some disadvantages, including poor adhesion because of low affinity with substrate surface and the need to use an adhesive layer due to evaporated temperature, metal and low melting point materials are suitable and refractory materials and multicomponent alloys cannot be used, contamination problems, and poor step coverage (rendering pattern making impossible). The film coated without contamination is made by materials that have a much higher vapor pressure than the heating element. The temperature and vapor pressure affect high deposition rates (up to  $100 \,\mu\text{m/min}$ ). Surface damage is low because the vapor particle energy is



FIGURE 7: Schematic of electron-beam evaporation process [41].

around 10~100 meV. The applications of DSSC are listed in Table 3.

Electron Beam Evaporation (EBE). Electron beam evaporation used high-speed electrons to bombard the source materials. During this process (Figure 7), the kinetic energy will be converted into thermal energy, which increases the temperature of the source. An electron gun creates the electron beam. The magnetic field bends at an angle of 270°, and the electron path bombards the position. The crucible acts as an anode and is cooled by cooling water to prevent contamination. Consequently, a pure thin film is formed. Quite a number of materials could be formed using this method, such as dielectric oxide, refractory metals, and metal oxide (Table 3). This method allows the easy control of the melting materials, which makes its deposition rate easily controlled as well  $(1 \text{ nm/min to } 1 \mu \text{m/min})$  using multiple guns and sources. These advantages enhance the uniform thickness and create a multilayer deposition by changing the crucible and making multiple target film possible. The surface, however, could easily be damaged by the decomposition or ionization of the target. However, the enormous electrical energy and Xray that is consumed in this method put it at a distinct disadvantage. The EBE used in DSSC application is shown in Table 4.

3.1.2. Sputtering. The sputtering method was discovered in 1852 as a means to monitor metals deposit at the cathodes of a cold cathode glow discharge. This method was used to coat mirror fabrics and phonograph wax masters. We analyzed two kinds of sputtering, including DC magnetron sputtering and radio frequency (RF) magnetron sputtering. DC magnetron sputtering could increase deposition rates with minimal damage to the substrate, while the RF magnetron sputtering plays an important role in allowing direct deposition of insulators. The high quality deposition materials such as metal, alloy, and simple organic compound coatings were deposited using the sputtering method, which

TABLE 4: List of materials by using EBE method in DSSC application.

Materials	Efficiency $\eta$ (%)	Reference
Ti electrode coated on TiO <sub>2</sub> layer	2.51	[116]
TiO <sub>2</sub> films	4.00	[117]
${\rm TiO_2}$ between the conductive transparent electrode and the porous ${\rm TiO_2}\mbox{-based}$ photoelectrode	7.01	[118]
Composite electrolyte (mixture of heteropoly acids (HPA) and polyethylene oxide (PEO))	3.40	[119]
TiO <sub>2</sub> film	6.64	[120]
Porous films of titanium oxide	4.10	[121]
Nanoporous films (TiO <sub>2</sub> -6 $\mu$ m)	6.10	[122]
Nanocolumnar titanium oxide films $(4  \mu m)$	—	[123]



FIGURE 8: Schematic of the processes that follow ion impact during sputtering [42].

precipitates the preference of this method over other PVD methods such as EBE and thermal evaporation (Table 5). Sputtering is considered a momentum transfer procedure. As the particles collide with a surface, the procedure is dependent upon the incident particle energy, the incidence angle, the surface atoms binding energy, and the colliding particles mass (Figure 8). The ions are regarded as the incident particles in sputtering, which are accelerated by an applied electrical potential. They will be reflected or absorbed by the surface when the kinetic energy is less than 5 eV. The surface will be damaged as the kinetic energy is higher than the surface atom binding energy when the atoms are placed into new lattice positions.

Sputtering occurs from the surface when the kinetic energy is above a certain threshold (10 to 30 eV). The sputtered atoms and ions deposit the thin film onto the substrate. A low-pressure process of the glow discharge type will usually cause energetic ion bombardment. The basic diode process is revealed in Figure 9. The vacuum chamber (argon, 10–3 to 10–1 torr) is supplied with a cathode as its target, the coating material source, and an anode as substrate for coating. During the sputtering process, the heat generated in the target is connected to a water-cooled metal (copper) backing plate with a solder or conductive epoxy, which cools the target. An electrical potential is applied between the cathode and anode. Grounded shields avoid discharges from forming in



FIGURE 9: Schematic of a diode sputtering assembly [43].



FIGURE 10: Graphic of DC and RF sputtering [44].

undesirable areas. The sputter yield is dependent on the target material; for example, silver has high yield. The deposition of large amounts of materials, including compounds and mixtures, the inducement of better step coverage and uniformity, and good adhesion with low substrate temperature are some of the advantages of this process, but the risk of substrate damage due to ionic bombardment is omnipresent. The list of materials used in DSSC application is shown in Table 4.

*DC Magnetron Sputtering.* DC magnetron sputtering is one of the simpler and older sputter depositions methods. It is made up of two parallel electrodes 4–8 cm apart, a cathode (target), and an anode (substrate) (Figure 10). The typical DC

Materials	Efficiency $\eta$ (%)	Reference
ITO-PEN	8.10	[94]
TiO <sub>2</sub> nanowires/nanoparticles hybrid membrane	2.50-2.70	[124]
ZnO nanostructured films	>4.58	[125]
Nanofibrous ZnO layers	1.70	[126]
ZnO compact layer	7.81	[127]
Pt counter electrodes on micropatterned ITO substrates	5.36	[128]
TiO <sub>2</sub> blocking-layer coated CdS-sensitized	0.52	[129]
PAMAM-like dendrons on SWCNT	_	[130]
TiO <sub>2</sub> microspheres with tunable sizes	6.78	[131]
Pt-counter electrode (Pt-CE)	7.54	[132]
Pt layer	5.11	[133]
Carbon films	5.00	[134]
Thin Pt electrode on transparent conductive oxide (TCO) substrate	6.00	[135]
Composite poly(3,3-diethyl-3,4-dihydro-2H-thieno-[3,4-b][1,4]-dioxepine) and platinum (PProDOT-Et <sub>2</sub> /Pt) film	6.68	[136]
Ultra-thin Pt films on indium tin oxide (ITO) substrates	7.90	[137]
ITO-x-SWCNT and ITO-x-SWCNT-N3 electrodes	_	[138]
${ m TiO}_2$ nanoparticles with natural dye extracted from pomegranate leaves and mulberry fruits	0.55-0.72	[139]
ZnO films of polyethylene glycol (PEG)	1.83	[140]
Pt/Ti bilayers	4.37	[141]
CdS-sensitized TiO <sub>2</sub> photoelectrode	1.04	[142]
Different TiO <sub>2</sub> passivating layers	2.85	[143]
ZnO nanorod	0.32	[144]
Ti doped ITiO films	3.80	[145]
FTO/nano-TiO <sub>2</sub>	4.40	[146]
FTO glass	11.18	[147]
ITO-PEN film coated Pt layer	5.24	[148]
Carbon electrode	3.89	[149]
Polymerized poly(3,4-ethylenedioxythiophene) on conductive glass	4.50	[150]

TABLE 5: DSSC application by using sputtering method	TABLE 5: DSS	C application	by using s	puttering	method
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potential that is applied is 1000–3000 V, the vacuum pump is  $10^{-3}$  Pa, and pressures are about 0.075 to 0.12 torr. Ions will be accelerated in the plasma to the cathode, then the target will be bombarded, and the sputtering energy is transferred from the target to the substrate's surface. This method should be used for conductive materials and is unsuitable for dielectric material targets (Table 6). The good adhesion, very low levels of contamination, film properties control of pressure, bias, and temperature are some of the advantages of DC sputtering, whereas some of its significant disadvantages include plasma electrons bombarding the substrate and producing heat, high working gas pressure, low deposition rate, and targets being limited to electrical conductors. The performance of this system can be improved by cathode systems.

*Radio Frequency (RF) Magnetron Sputtering*. Insulating materials are suitable as materials in RF sputtering, which are induced by a positive charge on the target surface (Table 7). The negative bias voltage is produced on the target when

the AC potential with high frequency is applied, mostly due to the mobility difference between electrons and ions. The positive ions bombard the target, leading to sputtering (Figure 10). The deposition rate of RF sputtering mirrors DC sputtering, where the target will be positive for a short time when the mass electron approaches the mass of ions. Therefore, the negative bias instigated sputtering on an insulating target. Practical applications utilized a frequency of around 13.56 MHz, and for industrial purposes, utilized a radio frequency designed by the Federal Communications Commission. RF sputtering deposits insulators with the same equipment and at lower pressures (5 to  $15 \times 10^{-3}$  torr), as well as conductors and semiconductors. The blockage of the RF radiation by electromagnetic shielding is one of the major disadvantages of RF sputtering. Additionally, resonant RF network requires complex equipment such as the power supplies and matching networks. Overall, RF sputtering demonstrates lower deposition rates compared to DC sputtering, but its damage to a substrate is significantly lower than that of its DC counterpart.

 TABLE 6: The list of materials used via DC magnetron sputtering in

 DSSC application.

Materials	Efficiency $\eta$ (%)	Reference
ZnO film	1.82	[151]
Pt metal grid	4.03	[152]
Compact TiO <sub>2</sub> deposited onto fluorine-doped tin oxide (FTO)	6.12	[153]
Pt/TiN thin film on PEN substrate	1.96	[154]
TiO <sub>2</sub> nanotube arrays	4.38	[155]
Nickel oxide (NiO <sub>x</sub> )	_	[156]
TiO <sub>2</sub> /NiO electrodes	4.16	[157]
TiO <sub>2</sub> nanorod arrays	4.78	[158]
Anatase TiO <sub>2</sub> thin films	_	[159]
Al-doped ZnO (AZO) films	0.61	[160]
Thin TiO <sub>2</sub> (anatase) films	_	[161]
TiO <sub>2</sub> films on SnO <sub>2</sub> :F coated glass substrates	0.78	[162]
Al <sub>2</sub> O <sub>3</sub> -coated TiO <sub>2</sub> electrode	5.91	[163]

TABLE 7: Materials in DSSC by RF magnetron sputtering methods.

Materials	Efficiency $\eta$ (%)	Reference
Aluminum-doped zinc oxide (AZO) thin films on glass substrates	2.91-4.13	[164]
Porous $TiO_2$ films on FTO electrode	4.60	[165]
Compact ZnO interlayer	4.00	[166]
TiO <sub>2</sub> covered FTO electrode	_	[167]
ZnO:Al thin film	2.10	[168]
Titanium-doped indium-tin-oxide (Ti:ITO) and indium-tin-oxide (ITO)	3.75	[169]
Indium tin oxide (ITO)	_	[170]
Aluminum oxynitride $(AlO_xN_y)$ films on polyethylene naphthalate (PEN) substrates	5.00	[171]
Triple-layered ITO/ATO/TiO <sub>2</sub>	6.29	[172]
Pt counter electrode	3.60	[173]
Columnar-structured rutile $TiO_2$ film	2.40	[174]

3.1.3. Pulsed Laser Deposition (PLD). The thin film deposition method used high power laser beams  $(108 \text{ W cm}^{-2})$ as a means of evaporation in ultra high vacuum chambers or gases such as oxygen (Figure 11). The laser is capable of evaporating the target source of heat or inducing photo dissociation. Then, the materials will be converted to plasma atoms, molecules, and chunks and are duly deposited onto the substrate. The film will not be damaged due to the absence of the accumulating of electric charge. The energy beam is regarded as a laser beam, which does not scatter for long distance. The target source decreases the contamination to the lowest level. This method is uncommon in the industry, due

to lasers being rather expensive. The advantage of this method is its alacrity and simplicity, that it maintains stoichiometry, that it is crystalline, its well-defined final structures, its high complexity and mixtures materials, its good adhesion, and its compatibility with oxygen and other reactive gases. All these properties could be controlled by laser intensity, substrate temperature, buffer gas pressure, and incident angle of the coating plume. Particulates, composition, and thickness being dependent on the deposition are some of the disadvantages of this process. Despite these considerable advantages, PLD is regarded as being too slow for industrial applications due to three main factors, such as the plasma forwarding directly to the substrate, being nonuniform, depositing in a relatively small area ( $\sim 1 \text{ cm}^2$ ), and requiring novel materials for the purpose of the optimization of deposition parameters. The novel PLD method called Glancing Angle Pulsed Laser Deposition (GAPLD) [49] is used to prepare porous nanostructure thin films. This process has the strength ability to control the shape of nanostructure, such as porous nanocolumnar film, nanorod arrays with different shapes, nanospring arrays, and multilayer nanostructures. In this method, the substrate is placed above the vapor source at an oblique angle. The composite TiO<sub>2</sub> electrodes show an efficiency of 3.80% [45], whereas the indium-tin oxide nanowires on glass have an efficiency of 0.15% [50]. The optical properties of ITO and TiO<sub>2</sub> single and double layer thin films developed the functional materials for DSSC applications [51], and the large surface area is imperative visà-vis dye-sensitized solar cells (DSSCs) applications [52].

3.1.4. Cathodic Arc Deposition (Arc-PVD). The arc-PVD is one of the ion beam deposition using electrical arc, which produces the blasts ions from the cathode (Figure 12). The high power density produced by the arc has a high level of ionization (30-100%), multiply charged ions, neutral particles, clusters, and macroparticles (droplets). The reactive gas is used throughout the evaporation procedure, and dissociation, ionization, and excitation could occur during the interaction with the ion flux, which results in a compound film being deposited. Metallic, ceramic, and composite films could be prepared with this method. The advantage of this process is its compatibility with industry, good adhesion of the product, stoichiometry compounds, low temperature, multilayer coating, compact film, uniform film, and low voltage. The inability to coat complex geometries and the utilization of liquid droplets and metal macroparticles are some of the disadvantages of this method [53]. Arc-PVD method is one the methods for fabrication DSSC such as ITO/Cr/DLC (n-type)/Pt (efficiency of 3.30%) [54], TiO<sub>2</sub>nanotube array (efficiency of 1.88%) [55], and nanocrystalline titania films (efficiency of 0.90%) [56].

3.1.5. Thermal Spraying (No High Vacuum). Thermal spraying is one of the coating methods that applies heat by electrical (plasma or arc) and chemical (combustion flame) means to melt the materials with high temperature and spray particles at high speeds on the surface (Figure 13). The liquid material will almost immediately form a film. Thermal spraying



FIGURE 11: Schematic of pulsed laser deposition [45].



FIGURE 12: Graphic of arc-PVD [46].

included plasma spraying, detonation spraying, wire arc spraying, flame spraying, velocity oxy-fuel coating spraying (HVOF), warm spraying, and cold spraying. The thickness of the deposited films usually ranges from  $20 \,\mu$ m to several mm, which is indicative of a high deposition rate in a large area relative to other chemical and physical vapor deposition methods. Quite a number of materials are viable for this process, such as metals, alloys, ceramics, plastics, and composites. This process is considered rather efficient

in the context of DSSCs, comparable to the doctor blading method [57]. The wide range of materials and substrates, drying process occurring in the spraying step, the formation of the film at low temperatures, and its rather quick processing method are some of the advantages of this method. Meanwhile, some of its disadvantages are the difficulty in controlling the thickness and its relatively low adhesion onto the substrate. There are some examples in DSSC application such as nanocrystalline TiO<sub>2</sub> (efficiency of 2.40%) [58],



FIGURE 13: Schematic of thermal spraying [47].





 $TiO_2$  films on ITO-PET substrates (efficiency of 2.80%) [59], and nanoporous microstructure of  $TiO_2$  (efficiency of 7.10%) [57].

3.1.6. Nanoparticle Deposition System (NPDS). NPDS is a novel method that is used to deposit ceramic and metallic powders on substrates at room temperature, by spraying particles under low temperature and low vacuums, and accelerate particles to subsonic speeds (Figure 14) [48, 60]. A novel method was applied for the deposition of  $TiO_2$  powders using Nanoparticle Deposition System (NPDS) for a dyesensitized solar cell (DSSC). This method could overcome certain limitations in the aerosol deposition method (ADM)

and cold gas dynamic spray method (CGDS). The ceramic powder deposited on the substrate by ADM in vacuum condition and metallic powder deposition requires a high pressure of carrier gas using the CGDS method. The ADM method could not be used for metallic powder, due to its low particle velocity [61]. This method could cover a wide range of materials using the supersonic/subsonic nozzle and low-vacuum deposition conditions [60]. Some advantages for this novel process are its viability at room temperature, susceptibility to a variety of materials, high deposition rates, and green fabrication without wet chemicals and binders [62]. NPDS is used to fabricate DSSC, for instance,  $TiO_2$ powders on indium tin oxide (ITO) coated polyethylene terephthalate (PET) substrates (efficiency of 0.94%) [63], semiconducting  $\text{TiO}_2$  powder on indium tin oxide (ITO)coated glass (efficiency of 1.24%) [60], and crystalline  $\text{TiO}_2$ powders (efficiency of 2.80%) [48].

#### 4. Conclusions and Outlook

Taking into account the discussion at this point, one can conclude that the choice of method depends on parameters that are regarded as important by the researchers. For example, if film adhesion forms the primary theme of the research, then the thermal spraying and PVD-thermal evaporation methods are unsuitable. If a substrate with a complex shape is subjected to coating, then the spin coating, PVD-thermal evaporation, and Arc-PVD do not meet the proper criteria. In addition, the limitations of precursor's choice in PVD-thermal evaporation and DC Magnetron sputtering are rather obvious.

The deposition methods are dependent on a variety of consideration and numerous criteria, including thin film applications, material characteristics, and process technology. The best deposition lacks a clear guideline and has to take into account the limitations and advantages of different preparations and deposition technology in the context of the materials' and substrates' type, specified application, cost, and requested efficiency, which could assist the researcher(s) in selecting a more appropriate procedure for their respective research.

#### **Conflict of Interests**

The authors declare that there is no conflict of interests regarding the publication of this paper.

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