

MOS Electron Transport Layers as Solar Cells

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ABSTRACT

Ultrasonic spray pyrolysis technique emerges as the simplest and most feasible. For thin-film preparation, this method is superior to other techniques because it produces relatively homogeneous compositions, allows easy control of film thickness, and yields a fine and porous microstructure. The size of the nanoparticle prepared from various techniques is less than 80 nm and it has spherical in shape in nature. In this review, reported methods for synthesizing potential ETL materials are summarized, and an attempt is made to guide further research in this field. With concerted efforts, the ultrasonic spray pyrolysis technique could serve as a change agent in synthesizing efficient materials for solar cell applications and thus significantly enhance their efficiency.

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Introduction

The tinier you get the more intricate properties you explore which can be put into numerous potential applications. The first decade of the 20th century revolutionized the entire material sciences with the first observations and size measurements of nanoparticles. By definition, nanomaterials must have at least one dimension which is less than approximately 100 nm. Hence, most nanoscale materials are too small to be visible to the naked eye or even conventional lab microscopes. However, owing to their size and morphology, nanomaterials can take on unique optical, magnetic, electrical, and other properties, thereby making their way into electronics such as photovoltaics and sensors, medicine, and other fields [1].

Metals can form several oxides with varied geometry and these oxides can exhibit metallic, semiconducting, or insulating character based on their electronic structure. In material engineering, oxides are used in the fabrication of microelectronic circuits, sensors, piezoelectric devices, fuel cells, coatings for the passivation of surfaces against corrosion, and as catalysts. Oxide nanoparticles can exhibit unique physical and chemical properties due to their limited size and a high density of corner or edge surface sites [2]. As a result, synthesis, characterization, and applications of metal oxide nanoparticles have attracted rapidly growing interest both for fundamental scientific interest and many practical and technological applications [3-9]. The shape control and assembly of nanostructures into organized patterns provide valuable routes to the design of functional materials and a variety of applications. Metal oxides nanoparticles such as TiO₂, ZnO, SnO₂, WO₃, In₂O₃, etc offer a wide range of applications [10-11]. For instance, TiO₂ can be used as a photocatalytic material [12], as a hydrogen gas sensor, as a solar cell material, etc; ZnO can be used in LPG sensing, solar cells [13-15], gas sensing [16], etc; SnO₂ can be

used for hydrogen gas sensing [17], in solar cells [15], etc; WO₃ can be used for electrochromic applications, solar cells, etc; and In₂O₃ can be used for solar cells [18,15], as hydrogen gas sensors [19], etc. The synthetic routes for the preparation of such inorganic nanomaterials can be broadly classified into two main groups - the “dry” methods and the “wet” methods. To obtain multifunctional inorganic nanoparticles, semiconductors, and quantum dots, with the desired size and morphology, it is necessary to optimize the parameters and develop the corresponding methods of synthesis. For the synthesis of nanoparticles ranging from micrometer to nanometer dimensions in powders, suspensions, or colloidal forms, a lot of different “wet” synthetic methods have been used: sol-gel, coprecipitation, microwave hydro (solvo) thermal, molten salt, polyol method, ultrasonic spray pyrolysis, organometallic, reverse-micelle, and colloidal chemical route, while the most preferred “dry” method is solid-state synthesis at high temperatures [20].

The sol-gel method can be understood as a two-step process - (i) hydrolysis of the precursor in the acidic or basic mediums and (ii) polycondensation of the hydrolyzed products. This method offers the advantage of obtaining a high surface area and stable surfaces [21]. On the other hand, co-precipitation is a simple yet effective method to prepare flowable powders without the need for any additional agglomeration steps. Moreover, it is an economical and industrially viable technique [22]. Coming to the hydrothermal method, it is a promising liquid phase preparation technology extensively used in the synthesis of a wide range of materials. Adding new force fields to this method results in several new methods such as the use of direct current, electric, magnetic, and microwave fields. The microwave hydrothermal method compensates for the lack of temperature in the hydrothermal method by employing the microwave temperature [23-24].

Most of these “wet” synthetic methods can produce powders with different morphology and varied sizes. These deposition techniques come with technical limitations such as compatibility

with large-area substrates, high cost, and need of high or sometimes even ultra-high vacuum [25]. However, the crystallinity of these nanoparticles needs to be improved by annealing the obtained samples. On the other hand, the process of ultrasonic spray pyrolysis can form fairly uniform, no agglomerated, and very porous submicron particles. Spray pyrolysis method is based on the formation of an aerosol from various precursor solutions, which could be a solution of metallic salts or a colloidal solution. The generated solution droplets also are known as the aerosol are then heated in a furnace very rapidly at a given temperature, and it passes through different stages - (1) evaporation of the solvent from the surface of the droplets, (2) drying the droplets containing the precipitated solute, (3) the annealing of the precipitate at high temperatures (thermolysis), (4) formation of microporous particles of defined phase composition, (5) formation of solid particles, and (6) sintering of solid particles. Each of these stages can be very conveniently tuned according to the final physical and chemical properties of the material in focus. The selection of both the precursor materials as well as the solvent affects the reaction parameters and hence the outcome of the process. Based on the requirement, thorough research has to be done to finalize the set of materials to be used [20, 26-30].

This method of Ultrasonic Spray Pyrolysis enables a fine mist of the precursor solution to deposit thin films of higher-density. Consequently, the use of this method for deposition of semiconductor thin films turns out to be low-cost, transparent, and flexible having large scale applications. Metal oxide semiconductors are very promising materials to be used in various electronic applications such as thin-film transistors (TFTs), sensors, electronic circuits, etc [25, 31-33]. The main limitation as per the reports is the need to use high deposition temperatures to achieve high-quality films as low temperatures often led to incomplete pyrolysis of the precursor solutions [34]. This issue can be resolved by the novel method of ultrasonic spray pyrolysis. Here, in this review, the application of this method is the synthesis of metal oxide nanoparticles for perovskite solar cells has been discussed. Though perovskite solar cells were well-known for years, the first incorporation was in 2009 by Tsutomu Miyasaka et al [35]. And over the years the efficiency of perovskite solar cells has gradually increased from 3.8% in 2009 [35] to 25.2% in 2020 in single-junction architectures [36]. and in silicon-based tandem cells, to 29.1% [36]. Currently, it is the fastest-growing solar cell technology with immense potential of high efficiency and low cost. The main use of metal oxide nanoparticles in these perovskite solar cells is as an Electron Transport Material, where

their major work is to prevent the recombination at the interfaces. These materials should be resilient to heat, light, and moisture to ensure the long-term stability of these perovskite solar cells. Metal oxides are best-suited for this purpose owing to their low-cost fabrication, long-term stability, and high efficiency [37]. Hence, the use of these metal oxides as Electron Transport Layer (ETL) has been elaborately discussed in this review along with the reported synthesis procedures for the same. Following this, an insight into the scope of future research is also provided.

Reported Experimental Section

Ultrasonic spray pyrolysis, owing to its easy feasibility, flexibility, and cost-efficiency, has gained popularity and is being extensively employed in the synthesis of nanomaterials such as thin films or nanoparticles. The precursors involved are often prepared by sol-gel methods [38]. Ultrasonic spray pyrolysis as a nano powder process belongs to a group of solution aerosol thermolysis, which is a general group of processes for particle production which are based on precursor atomization (droplet size 1-100 micrometer), aerosol transport through a temperature and atmosphere regulated reactor. Particle morphology in these processes is the result of droplet size, precursor concentration, and physical characteristics, operating temperature, and evaporation rate [39].

The typical spray pyrolysis apparatus consists of an atomizer, precursor solution, substrate heater, and temperature controller. There are two different ways this method is employed - (i) powder preparation and (ii) thin film preparation.

Powder Preparation

For the preparation of powder using the ultrasonic spray pyrolysis technique, the major components are - (i) the ultrasonic atomizer with three transducers for conversion of precursor solution into aerosol, (ii) the quartz reactor placed in a two-zone pyrolysis furnace, and (iii) novel trapping system to collect the nanocrystalline powder. After the aerosol formation, it is pushed forward through the quartz reactor using compressed air or pure oxygen. The aerosol would have to pass through multiple processes in sequence to finally form the fine particles. Some of these processes include heating, solvent evaporation, and pyrolysis reaction with air/oxygen. A novel trapping system can be employed to sort the nanoparticles size-wise [40].

A simple schematic diagram of the process has been shown below in Figure 1 [40].

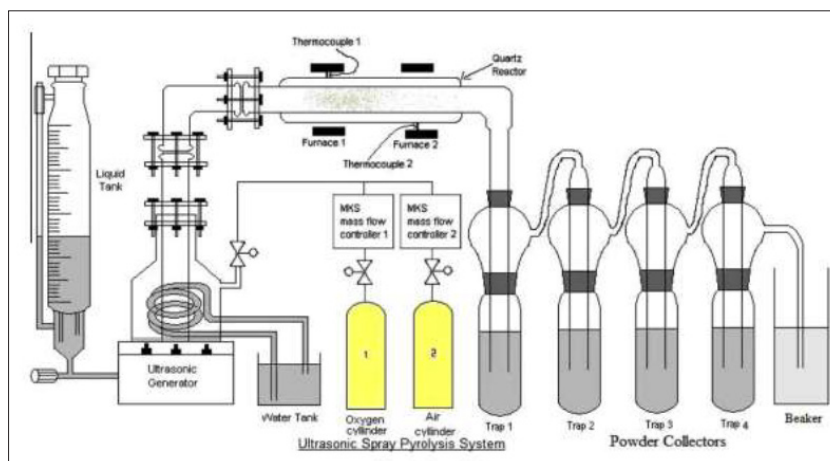


Figure 1: A Schematic Diagram for the Powder Synthesis using the Ultrasonic Spray Pyrolysis Method [40].

Thin Films Preparation

The method of ultrasonic spray pyrolysis weighs much above other methods of synthesis of thin films because of relatively homogeneous composition, easy control of film thickness, and fine and porous microstructure apart from the factors like low cost, suitable properties and well-suited process to large-scale production. The simple set-up consists of a spraying chamber, ultrasonic generator, spray nozzle (Sono-Tek) attached to the x-y plotter, syringe pump, heating system with bronze block, and temperature indicator. The Ultrasonic generator produces high frequency (120 kHz) ultrasonic waves, which when applied to the piezoelectric transducer, mechanical vibration is generated. These vibrations convert the precursor solution into the spray (mist) and this spray is then used to prepare nanocrystalline thin films [14]. Few problems encountered in sprayed films are due to non-uniform droplet sizes formed at the outlet of the nozzle. This results in poor grain size and hence lower transparency. Lampkin [41]. through a thorough investigation of the aerodynamics of the spray nozzle and Som and Mukharjee [42]. through a study on the discharge coefficient and spray cone angle of the swirling spray atomizing nozzle, have treated this effect.

A simple schematic diagram of the process has been shown below in Figure 2 [14].

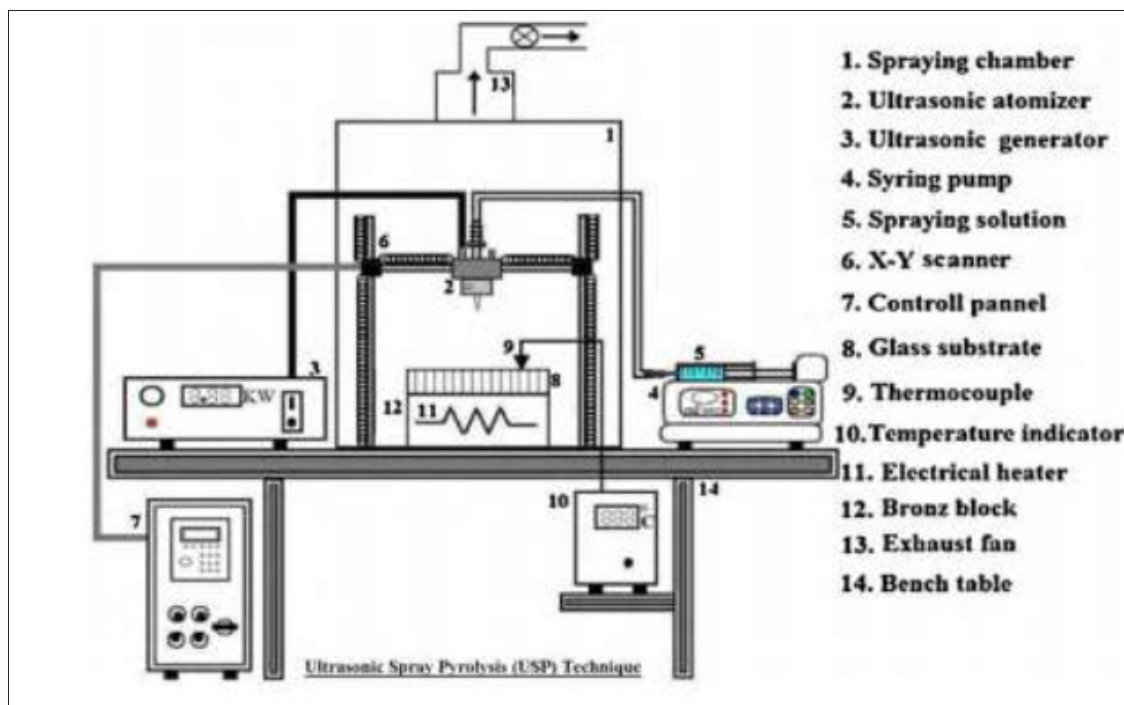


Figure 2: A Schematic Diagram for the Thin Film Synthesis Using the Ultrasonic Spray Pyrolysis Method [14].

Simple equipment and experimental arrangement have made this method cost-effective and the most significant factor is the extensive control over the morphology and size by varying the preparative conditions, nature of additives, flow rate, and concentration of reactants in the precursor solutions. Furthermore, there are continuous processes that assist in establishing better control over morphology and getting fine powders with spherical particles and the desired diameter dictated by the size of the drops. Like every other method, this method of spray pyrolysis also has some disadvantages like low yield, oxidation of sulphides during processing in air, and difficulties associated with growth temperature determination. However, the advantages overpower these demerits and have made this method a widely accepted one [20].

The spray pyrolysis method stands out as one of the most preferred methods through which porous films and films with high-density packaging and high uniformity of particles are obtained [43]. This method does not fail us even when it comes to the synthesis of ultrafine powders with small particle size ($< 1 \mu\text{m}$), narrow size distribution ($1\text{-}2 \mu\text{m}$), high purity, high porosity, and large surface area [44-49]. This method has been employed to deposit a broad range of thin films which have been used in various devices such as solar cells, sensors, and solid oxide fuel cells. A reactive spray technique has been used for long to prepare transparent conducting oxide layers [50-57], but considerable efforts to produce high-quality oxide films such as SnO_2 , CdSnO_4 , In_2O_3 , TiO_2 , etc [58-63]. has been given around mid-seventies because of their applications in solar cells and gas sensing elements. In this review, we will attempt to highlight the use of ultrasonic spray pyrolysis technique in the synthesis of metal oxides that work as Electron Transport Layers (ETLs) in perovskite solar cells [64-65].

Electron Transport Layer (ETL)

The emergence of the perovskite solar cells brings with it a new revolution in the solar cell industry owing to the incredible improvement of their conversion efficiency, which can exceed 20.0% within seven years of extensive research with economic production costs. Many oxide semiconductors have been reported to enhance the stability of these solar cells. Semiconductors used as transparent electrodes include oxides such as tin oxide, indium oxide, cadmium oxide, zinc oxide, etc. Some of the methods used to deposit these semiconductors in the form of thin films include - R.F. sputtering [66-68]. D.C. diode sputtering [66]. reactive sputtering in O_2 [69-70].

chemical vapor deposition [71-72]. oxidation under vacuum [73-74]. flash evaporation [75]. and spray pyrolysis [75-86]. Among all these methods, spray pyrolysis is the most preferable method for the deposition of thin films.

The efficiency and stability of these perovskite solar cells heavily depend on the morphology and type of materials opted for as the Electron Transport Layer (ETL) in the device. Before we move on to selecting the kind of materials to use, it is necessary to understand the role of an ETL and the types of device architecture that exist. The presence of an ETL is essential to facilitate efficient electron collection and transportation from the perovskite layer to its respective electrodes. The absence of ETL has a very small impact on the open-circuit voltage (VOC), but it drastically reduces the short-circuit current density (JSC) as well as the fill factor (FF). This in turn affects the conversion efficiency of the devices. Coming to the different types of device architecture that exist, there are two types - planar and mesoporous configuration [87]. The mesoporous layer is just an optional layer employed to improve on the electron extraction efficiency and minimize the hysteresis effect. Using a compact ETL in the device is extremely crucial as it prevents the charge recombination at the interface [87-89]. A clearer picture of the device architecture has been shown below in Figure 3.

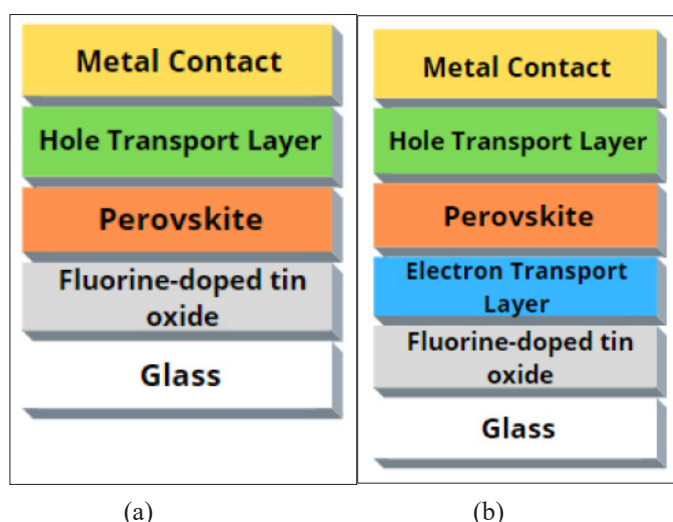


Figure 3: The Device Architecture of the Perovskite Solar Cells (PSC) - (a) the Planar PSC without the Electron Transport Layer (ETL) and (b) the Planar PSC with the Electron Transport Layer (ETL)

The compact ETL in a planar heterojunction perovskite solar cell has multiple roles like ensuring an easy and smooth transition of the photoexcited electrons from the perovskite active layer to the external circuit and acting as a hole blocking layer (HBL) to suppress the transportation of the photogenerated holes from the perovskite light-harvester to the FTO substrate. The ETL needs to have a continuous, uniform, and ultrathin structure [87].

Most of the reviews in this field cover aspects of metal oxide Electron Transport Materials (ETMs), comparisons between compact and mesoscopic structures, effects of ETMs on both device efficiency and stability [90-101]. In this review, an attempt has been made to elaborate on the role of ETL and provide a succinct review of the metal oxides that can be used as ETL materials. Here, TiO₂, ZnO, SnO₂, WO₃, and In₂O₃ have been taken into consideration. A summary of the properties of these oxides like mobility, conduction band minimum (CBM), and

valence band maximum (VBM) have been given below in Table 1.

Table 1: Common Properties of ETL Materials

Sr. No	ETM	Mobility (cm ² V ⁻¹ s ⁻¹)	CBM (eV)	VBM (eV)	References
1	TiO ₂	0.1-4	- 4.1 (anatase) - 3.9 (rutile) - 4.1 (brookite)	- 7.3 - 6.9 - 7.4	[93] [94] [95]
2	ZnO	205-300	- 4.2	- 7.6	[96-97]
3	SnO ₂	240	- 4.5	- 8.1	[98-99]
4	WO ₃	-	- 5.3	- 8.3	[100]
5	In ₂ O ₃	66	- 0.37	-3.0	[101]

Thermodynamic considerations show that the VBM position is suitable for hole separation and the CBM position is suitable for electron separation. And based on bandgap energy, the absorption onset wavelength can be predicted [102]. It has been stated that a metal oxide with high electron mobility might improve the photovoltaic performance [103]. Hence, ZnO and SnO₂ stand out as potential candidates for consideration in solar cell applications.

Now that we understand the role that ETL plays in a perovskite solar cell, it will be easier for us to develop the set of criteria essential for a material to qualify as a good ETL material. Some of the qualities that a good ETL material must possess (1) good electron mobility to help in fast transport of electrons within the ETL, (2) a wide bandgap to achieve high transparency to maximize light absorption by the perovskite materials, and (3) compatible energy levels - compatible conduction band minimum (CBM) and valence band maximum (VBM) as compared to the energy levels of the perovskite light-harvester. A very wellknown material used in the ETL in perovskite solar cells is TiO₂. It has an electron mobility of approximately 1 cm²V⁻¹s⁻¹ with a CBM of -4.2 eV vs. a vacuum level. The CBM of TiO₂ is relatively 0.3 eV lower than that of the MAPbI₃ perovskite so that the injection of photoelectrons from the perovskite material to ETL is facilitated. Even the VBM of TiO₂ is much lower than the perovskite materials, thereby enhancing the hole-blocking trait. Generally, the perovskite solar cells face stability issues as they are affected by UV light and moisture. It has been reported that TiO₂-based perovskite solar cells show instability under UV illumination due to the light-induced desorption of surface-adsorbed oxygen in TiO₂ [104]. For avoiding damage due to moisture, one can opt for mesoporous ETL, and for dealing with the UV light issue, one needs to use materials that are insensitive to UV radiation. Other materials which can substitute TiO₂ are ZnO, SnO₂, WO₃, In₂O₃ etc [87, 105-107].

Spray pyrolysis method has proved its worth as a deposition method in the production of high-efficiency solar cells in a relatively simple, fast, and inexpensive way without the need for vacuum or the use of high-temperature processes. A detailed description of the synthesis method for some of these oxides for the applications in solar cells using Ultrasonic Spray Pyrolysis technique has been discussed below:

Synthesis of Metal Oxide Thin Films ZnO Thin Films

Zinc oxide crystallizes in the wurtzite structure and can be

synthesized in the form of thin films with degenerate n-type doping. It has a very high bandgap (~ 3.2 eV), which makes it suitable for transparent electrode applications. Thin films of ZnO with high transparency and low resistivity can be produced by spray pyrolysis method with an aqueous solution of a zinc salt as a starting material and then spraying it over a heated substrate. Conventionally, solutions containing Zn (NO₃)₂, Zn(C₂H₃O₂)₂, and ZnCl₂ are put into use. For films made from ZnCl₂ precursor solutions, there is a significant decrease in the Cl concentration with substrate temperature and airflow rate in the sprayer. This clearly explains the necessity of high substrate temperatures to obtain high transparency. The optical transmission of the films could also be enhanced by using hydrogen peroxide to the starting precursor solution because hydrogen peroxide reduces the amount of Cl in the film by forming volatile compounds. In cases of using Zn(C₂H₃O₂)₂ and ZnCl₂ as starting materials, a 420°C substrate temperature has been found to give minimum resistivity [108].

SnO₂ Thin Films

SnO₂ offers better chemical stability and the possibility of modifying the alloy composition, thereby allowing certain amplitudes in the electrical properties of the material. Tin oxide is an n-type semiconductor with a tetragonal crystal structure and a wide bandgap (~ 3.5 eV). The deposition of tin oxide films can be employed by spraying a hydroalcoholic solution of tin tetrachloride along with a suitable dopant if needed onto a heated substrate. The following chemical reaction summarizes the process –



This endothermic reaction takes place at the surface of the substrate. To lower the resistivity of the thin films, fluor can be added to the solution in the form of trifluoroacetic acid or ammonium fluoride. The optimum substrate temperature for the reaction is 500°C. Typical spray flow rates offer fast deposition rates up to 0.2 to 0.4 µm/min. The deposited tin oxide films have granular polycrystallinity with a tetragonal rutile structure [109].

In₂O₃ Thin Films

In₂O₃ is also an n-type oxide semiconductor with a wide bandgap and degenerate doping.

The reaction occurring at the substrate surface is –



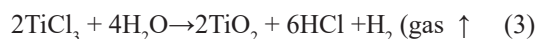
For spraying this oxide semiconductor, usually, indium oxide is mixed with tin oxide in varying proportions to improve the electrical conductivity of the resulting films. For solar cell applications, higher concentrations and lower substrate temperatures (~ 440°C) are advisable [15].

Reports on the use of Ultrasonic Spray Pyrolysis for the synthesis of metal oxides like TiO₂ and WO₃ for applications in solar cells could not be discovered in our search. Hence, different reported synthesis methods of these metal oxides for other applications have been discussed.

TiO₂ Thin Films

For applications such as gas sensing, TiO₂ thin films are synthesized using TiCl₃ (0.05 M) as the precursor. The solution is sprayed onto a glass substrate heated at 350°C and then fired for one hour at 550°C [13].

The reaction occurring at the substrate surface is -



For applications such as photocatalytic materials, the precursor used is different. The spray solution is made up of titanium (IV) isopropoxide (0.2 mol/L) and acetylacetone in a molar ratio of 1:4 in ethanol and the spraying rate is set up at 2.5 mL/min. Compressed air is also applied at the flow rate of 8 L/min as the carrier gas. The hot plate temperature is adjusted to 250°C, 350°C and 450°C, following which all the samples are annealed at 500°C. The obtained films are smooth, dense, and free of cracks. All the obtained films possess an anatase crystalline structure. However, with the increasing temperature of the hot plate, the surface morphology changes, and an increase in the film thickness is observed [12].

Synthesis Method for Wo₃ Thin Films

Thin films of tungsten oxide along with growth controlling agents such as hexadecyltrimethylammonium bromide (HTAB) and polyethylene glycol (PEG400) have been reported. The precursor used was tungsten chloride (WCl₆) powder in ethanol containing acetylacetone (AcAc) with WCl₆ and AcAc in a 1:2 M ratio. HTAB and PEG400 were also added to the spraying solution. The deposition was done at a substrate temperature of 250°C, airflow pressure of 150 kPa, the spraying angle as 45°, and the spraying distance as 23 cm. Fifteen consecutive layers were deposited with 100s break in between and the samples were annealed for six hours at 410°C. Very smooth and homogeneous layers with the monoclinic structure were obtained [18].

Comparative study of Reported Metal Oxide Thin Films

The use of different metal oxides (including dopants), their respective processing parameters, and solar cell efficiency has been summarized below in Table 2.

- a. NR = Not Reported
- b. HTAB = Hexadecyltrimethylammonium Bromide
- c. PEG400 = Polyethylene Glycol
- d. Illumination of 25 mW/cm²
- e. Illumination of 80 mW/cm²

Table 2: Different Metal Oxides Thin Films Preparation and Their Applications

Sl. No	Metal oxide	Dopant	Solution	Tsubstrate (°C)	Thickness (μm)	Area (cm ²)	Applications	Solar Cell Efficiency(%)	Year	Ref
1	SnO ₂	NR ^a	0.77 M of SnCl ₄ in ethyl acetate	0.77 M of SnCl ₄ in ethyl acetate	0.1(Single Si)	3.8	Solar Cells	13.60	1979	[80]
2	SnO ₂	NR	0.77 M of SnCl ₄ in ethyl acetate	300 - 400	0.1(Single Si)	20	Solar Cells	~10	1979	[80]
3	SnO ₂	NR	0.77 M of SnCl ₄ in ethyl acetate	300 - 400	0.1(Poly-Si)	1	Solar Cells	12.20	1979	[80]
4	SnO ₂	NR	An alcoholic solution of SnCl ₄ .H ₂ O	600	0.1(Single Si)	0.3d	Solar Cells	6.7	1979	[82]
5	SnO ₂	NR	An alcoholic solution of SnCl ₄ .H ₂ O	600	0.1(Single Si)	0.3e	Solar Cells	7.2	1979	[82]
6	In ₂ O ₃	2%SnCl ₄ .5H ₂ O	19%InCl ₃ + 41% H ₂ O+40% C ₂ H ₅ OH	500	0.24(Single Si)	1	Solar Cells	11.31	1979	[78]
7	In ₂ O ₃	2%SnCl ₄ .5H ₂ O	19%InCl ₃ + 41% H ₂ O+ 40% C ₂ H ₅ OH	500	0.24(Poly-Si)	1	Solar Cells	9.1	1979	[78]
8	In ₂ O ₃ [84]	0.25 Sn/In atomic ratio	An alcoholic solution of SnCl ₄ .H ₂ O +InCl ₃	435	NR(Single Si)	0.1	Solar Cells	10.2	1977	[84]
9	In ₂ O ₃	0.25 Sn/In atomic ratio	An alcoholic solution of SnCl ₄ .H ₂ O +InCl ₃	435	NR(Single Si)	1.5	Solar Cells	9.1	1977	[84]
10	In ₂ O ₃	2.5 SnCl ₄ .5H ₂ O concen ring InCl ₃	Equal proportions of ethyl alcohol +water+(InCl ₃ + SnCl ₄ .H ₂ O	400	0.3 - 0.4 (Single Si)	0.3	Solar Cells	11.5	1980	[86] [108]
11	ZnO	NR	Zinc chloride or Zinc acetate	460	0.5(Single CdTe)	0.17	Solar Cells	8.8	1980	[109]
12	TiO ₂	NR	Titanium (IV) isopropoxide: acetylacetone = 1:4 in ethanol	(i)250 (ii)350 (iii)450	(i)0.11 (ii)0.18 (iii)0.24	NR	Photocatalytic material for air purification	NR	2019	[12]
13	WO ₃	NR	WCl ₆ :Acetylacetone = 1:2 in ethanol + HTABband PEG400c	250	0.2	NR	Electrochromic applications	NR	2013	[18]

- NR = Not Reported
- HTAB = Hexadecyltrimethylammonium Bromide
- PEG400 = Polyethylene Glycol
- Illumination of 25 mW/cm²
- Illumination of 80 mW/cm²

Based on a recent report on the choice of ETL materials, instead of the conventional TiO₂ or ZnO layer, WO₃ has been considered as a potential candidate for the Electron Transport Layer with an efficiency of 15.54% under optimal conditions [110]. It is cost-effective and easy to fabricate [111].

Remarks

Among the various reported methods of synthesis of nanomaterials and putting them into use in different applications, Ultrasonic Spray Pyrolysis is considered to be one of the easiest, feasible, and cost-effective methods. The simple equipment and experimental arrangement add to the feasibility of the technique and the most important factor that this technique offers is extensive control over the morphology and size by slight modifications of the preparative conditions, nature of additives, flow rate, and concentration of reactants in the precursor solutions. Also, this method proceeds via various continuous processes that support in establishing better control over morphology and getting fine powders with spherical particles and the desired diameter dictated by the drop-size. Even though this method provides low yield and has issues like oxidation of sulphides during processing in air and difficulties in growth temperature determination, it stands out as one of the most preferred methods [20].

Coming to the preparation of thin films, this method helps to obtain porous films and films with high-density packaging and high uniformity of particles [43]. It also assists in the synthesis of ultrafine powders with small particle size ($< 1\ \mu\text{m}$), narrow size distribution ($1\text{--}2\ \mu\text{m}$), high purity, high porosity, and large surface area [44–49]. Because of relatively homogeneous composition, easy control of film thickness and fine and porous microstructure, this method is extensively employed in the synthesis of thin films which have been used in various devices such as solar cells, sensors, and solid oxide fuel cells. This technique of Ultrasonic Spray Pyrolysis is our method to synthesize metal oxide thin films for use as ETL materials in perovskite solar cells. The heavy dependence of the efficiency and stability of the perovskite solar cells on the morphology and type of ETL material used in the device makes it extremely necessary to choose the right material. The ETL material in a device facilitates efficient electron collection and transportation from the perovskite layer to its respective electrodes. The absence of the ETL layer affects the performance of the device. Hence, using a compact ETL is extremely essential to prevent the charge recombination at the interface [87–89]. The ETL must have a continuous, uniform, and ultrathin structure [87]. Based on the list of criteria for a substance to be a good ETL material, TiO_2 , with electron mobility of approximately $1\ \text{cm}^2\text{V}^{-1}\text{s}^{-1}$ with a CBM of $-4.2\ \text{eV}$ vs. vacuum level, qualifies as a good candidate. However, there are stability issues in the presence of UV light and moisture. For avoiding damage due to moisture, one can opt for mesoporous ETL, and for dealing with the UV light issue, one needs to use materials that are insensitive to UV radiation. Other materials that can substitute TiO_2 are ZnO , SnO_2 , WO_3 , In_2O_3 , etc [87]. Synthesis methods for these metal oxides have been elaborated in the above section.

From Table 2, it can be observed that when the area decreases, the solar cell efficiency of the device goes down. For a comparison, consider Sl. No. 1 and Sl. No. 2 from the table. While the other parameters are constant, when the area of the device decreases, the performance also decreases. Another striking observation would be the smaller efficiency in the case of Poly-Si as compared to that of Single Si. For this comparison, consider Sl. No. 6 and Sl. No. 7 from the table. And comparing the Sl. No. 4 and Sl. No. 5 from the table, it can be concluded that greater illumination results in better efficiency of the device, and this indeed is completely expected. In the case of TiO_2 , the thickness of the films increases with increasing substrate temperature as shown in Sl. No. 12. This might be true in the case of other metal oxides as well, but experimental data is necessary to validate these expected results. It has been proposed to use WO_3 as an ETL material to obtain maximum efficiency.

Conclusion

Synthesis of metal oxides using the Ultrasonic Spray Pyrolysis technique for Electron Transport Layers in perovskite solar cells is an extremely successful approach and can solve the present limitations due to the other available methods of synthesis. Three of the five metal oxides considered for this review have already been synthesized using this technique and specifically for their application in solar cells. Numerous reports on this technique being widely employed for the synthesis of various materials have been considered for this review and the extensive use of the method clearly explains its superior nature as compared to the other methods of synthesis. Proper research efforts directed towards the synthesis of ETL materials using this technique can revolutionize the entire solar cells industry and can increase the efficiency of solar cell manifolds. This can act as the change needed and can heavily contribute to human society.

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Competing Interests

The authors hereby declare that there is no competing interest among the authors.

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