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# Perspective on Predominant Metal Oxide Charge Transporting Materials for High-Performance Perovskite Solar Cells

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Nowadays, the power conversion efficiency of organometallic mixed halide perovskite solar cells (PSCs) is beyond 25%. To fabricate highly efficient and stable PSCs, the performance of metal oxide charge transport layers (CTLs) is one of the key factors. The CTLs are employed in PSCs to separate the electrons and holes generated in the perovskite active layer, suppressing the charge recombination rate so that the charge collection efficiency can be increased at their respective electrodes. In general, engineering of metal oxide electron transport layers (ETLs) is found to be dominated in the research community to boost the performance of PSCs due to the resilient features of ETLs such as excellent electronic properties, high resistance to thermal temperature and moisture, ensuring good device stability as well as their high versatility in material preparation. The metal oxide hole transport layers in PSCs are recently intensively studied. The performance of PSCs is found to be very promising by using optimized hole transport materials. This review concisely discusses the evolution of some prevalent metal oxide charge transport materials (CTMs) including TiO<sub>2</sub>, SnO<sub>2</sub>, and NiO<sub>x</sub>, which are able to yield high-performance PSCs. The article begins with introducing the development trend of PSCs using different types of CTLs, pointing out the important criteria for metal oxides being effective CTLs, and then a variety of preparation methods for CTLs as employed by the community for high-performance PSCs are discussed. Finally, the challenges and prospects for future research direction toward scalable metal oxide CTM-based PSCs are delineated.

**Keywords:** metal oxides, electron transport layers, hole transport layers, cost-effective, stability, perovskite solar cells

## INTRODUCTION

In the 21st century, we are living in an energy-driven world. Sustainability in energy generation is one of the most challenging issues to satisfy the ever-increasing energy demands. Solar energy is readily available in nature, which exhibits a great potential to address the global energy challenge. However, the widespread use of solar energy requires the development of cost-effective and high-performance photovoltaics (PVs) for efficient energy conversion from solar to electric energy. The emergence of organometallic mixed halide perovskite solar cells (PSCs) resulted in an important

breakthrough in PV technology. The recently announced record power conversion efficiency (PCE) values of single-junction PSCs and perovskite-based monolithic tandem devices are 25.5 and 29.5%, respectively (The National Renewable Energy Laboratory [NREL], 2021), which outperform other thin-film PV technologies (e.g., CIGS and CdTe) and are comparable to well-established solar technologies such as crystalline silicon photovoltaics (PCE ~26.1%). Such impressive device efficiency of PSCs is owing to inherent perovskite properties such as large absorption coefficients, adjustable bandgaps, low exciton binding energy, large carrier diffusion lengths, and high charge mobilities (Xing et al., 2013; Wang et al., 2015). However, a number of challenges such as device long-term stability, hysteresis effect, toxicity of lead, and scalable difficulties still remain in the community (Djurišić et al., 2016; Djurišić et al., 2017; Li et al., 2018; Li et al., 2020). In general, five major areas are being investigated to come up with possible solutions for the problems mentioned above. They include (i) perovskite composition and crystal structure (Luo and Daoud, 2016; Mitchell et al., 2017), (ii) engineering of perovskite growth methods (Ng et al., 2015, 2018; Babayigit et al., 2018; Shen et al., 2018; Hu et al., 2020a), (iii) optimization of carrier transport materials and interfaces (Liu et al., 2017; Hu et al., 2020b,c; Wang et al., 2021), (iv) device architectures (Hanmandlu et al., 2018; Ren et al., 2018; Maxim et al., 2020; Shalenov et al., 2020), and (v) encapsulation (Dong et al., 2016; Fu et al., 2019; Emami et al., 2020). Some comprehensive review papers on those topics have also been published (Jiang et al., 2018; Sajid et al., 2018; Ouyang et al., 2019; Hanmandlu et al., 2020; Hu et al., 2020a; Zhou et al., 2020).

This article aims to highlight the development of the predominant metal oxides—TiO<sub>2</sub>, SnO<sub>2</sub>, and NiO<sub>x</sub>—using the charge transporting layers (CTLs) in PSCs. Special attentions are placed on these three types of metal oxides as the state-of-the-art PSCs with record-breaking efficiencies are usually associated with these metal oxide CTLs. A brief review on representative research works on PSCs has been done, indicating the highest achievable PCE for different CTL-based PSCs at different stages of time. The important criteria in selecting metal oxides as CTLs are emphasized. Varieties of effective preparation methods for metal oxide CTLs have been summarized, and their advantages and shortcomings in terms of processing conditions and possibility of future large-scale manufacturing are discussed. This work will provide a direct insight into the PV community for further optimizing of CTLs, which will be one of the critical steps for approaching practical PSCs in the commercial market.

## THE EFFICIENCY EVOLUTION OF PSCS USING DIFFERENT METAL OXIDE CTLs

Starting from 2009, the group of Miyasaka demonstrated the photovoltaic properties of using perovskite in the dye-sensitized solar cell by employing mesoporous TiO<sub>2</sub>, exhibiting a PCE of 3.8% (Kojima et al., 2009). In early 2012, the group of Park reported a PCE of 9.7% of PSCs by introducing a sub-micrometer-thick layer of TiO<sub>2</sub> and solid-state hole transporting materials (HTMs) (Kim et al., 2012). In the

same year, a PCE more than 10% was reported by the group of Snaith (Lee et al., 2012). In 2013, the group of Grätzel demonstrated high-performance PSCs fabricated by the sequential deposition method with a certified PCE of 14.1% (Burschka et al., 2013). In the following years, the group of Seok reported a dramatical improvement in the device performance of PSCs by using different engineering approaches such as solvent engineering (certified as 16.20%) (Jeon et al., 2014), compositional engineering (certified as 17.9%) (Jeon et al., 2015), and intramolecular exchange (20.1% certified PCE) (Yang et al., 2015c). In 2016, the group of Grätzel reported the use of triple cations in perovskite materials, yielding good device reproducibility and improved device stability, leading to a high PCE of 21.1% (Saliba et al., 2016). Nowadays, the highest certified PCE as reported in literature for PSCs based on TiO<sub>2</sub> electron-transporting layer (ETL) is 24.64% (Jeong et al., 2020). The TiO<sub>2</sub>-based ETLs are popular to be used since the early stage of PSCs owing to its high versatility of preparation techniques as well as attributing to the evolution of the architecture from the dye-sensitized solar cells. The representative research works using TiO<sub>2</sub> as the ETL in PSCs with their achieved PCEs are summarized in **Figure 1** as indicated in solid circles. Along with such a tremendous improvement in device performance, a compact layer of tin oxide (SnO<sub>2</sub>) has been demonstrated as an alternative for ETL. First, Ma et al., 2017 reported a high-temperature processing compact SnO<sub>2</sub> ETL prepared by the conversion of SnCl<sub>2</sub>·2H<sub>2</sub>O precursor to SnO<sub>2</sub> using a sol-gel method for CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>-based PSCs, showing a PCE of 7.43% (Dong et al., 2015). The group of Dai and Kuang also reported a high-temperature processing SnO<sub>2</sub> mesoporous ETL and as-synthesized colloidal solution, respectively, followed by TiCl<sub>4</sub> surface treatment for CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>-based PSCs, exhibiting an improvement in their champion device efficiency of 10.18 (Li et al., 2015) and 14.69% (Rao et al., 2015), respectively. However, at that time, the PCEs of SnO<sub>2</sub>-based PSCs are still lower than those of TiO<sub>2</sub>-based PSCs due to the introduction of a large amount of trap states during the high-temperature processing (Ke et al., 2015). The important breakthrough in achieving high device efficiency for SnO<sub>2</sub>-based PSCs was from 2015. The group of Fang firstly introduced low-temperature processing for preparing SnO<sub>2</sub> compact layer based on the sol-gel method using SnCl<sub>2</sub>·2H<sub>2</sub>O as the precursor to achieve a high PCE of 17.21% for CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub>-based PSCs (Ke et al., 2015). Then, the group of You reported a PCE of 20.54% (certified as 19.9%), which was a record-high value in 2016 for (FAPbI<sub>3</sub>)<sub>x</sub>(MAPbBr<sub>3</sub>)<sub>1-x</sub>-based PSCs with a low-temperature processing SnO<sub>2</sub> ETL prepared from commercially available SnO<sub>2</sub> colloid solution (Jiang et al., 2016). In the same year, the group of Hagfeldt demonstrated a low-temperature chemical bath deposition for preparing SnO<sub>2</sub> compact layer as ETL used in Cs-containing mixed halide-based PSCs, yielding a stabilized PCE of 20.7% (Anaraki et al., 2016). In 2017, the group of You updated the champion PCE to 21.6% (certified as 20.9%) using low-temperature processing SnO<sub>2</sub> in (FAPbI<sub>3</sub>)<sub>1-x</sub>(MAPbBr<sub>3</sub>)<sub>x</sub>-based PSCs (Jiang et al., 2017). The representative research works using SnO<sub>2</sub> as the ETL in PSCs with their achieved PCEs are summarized in **Figure 1** as indicated in solid squares. On the other hand, metal oxides based on NiO<sub>x</sub>

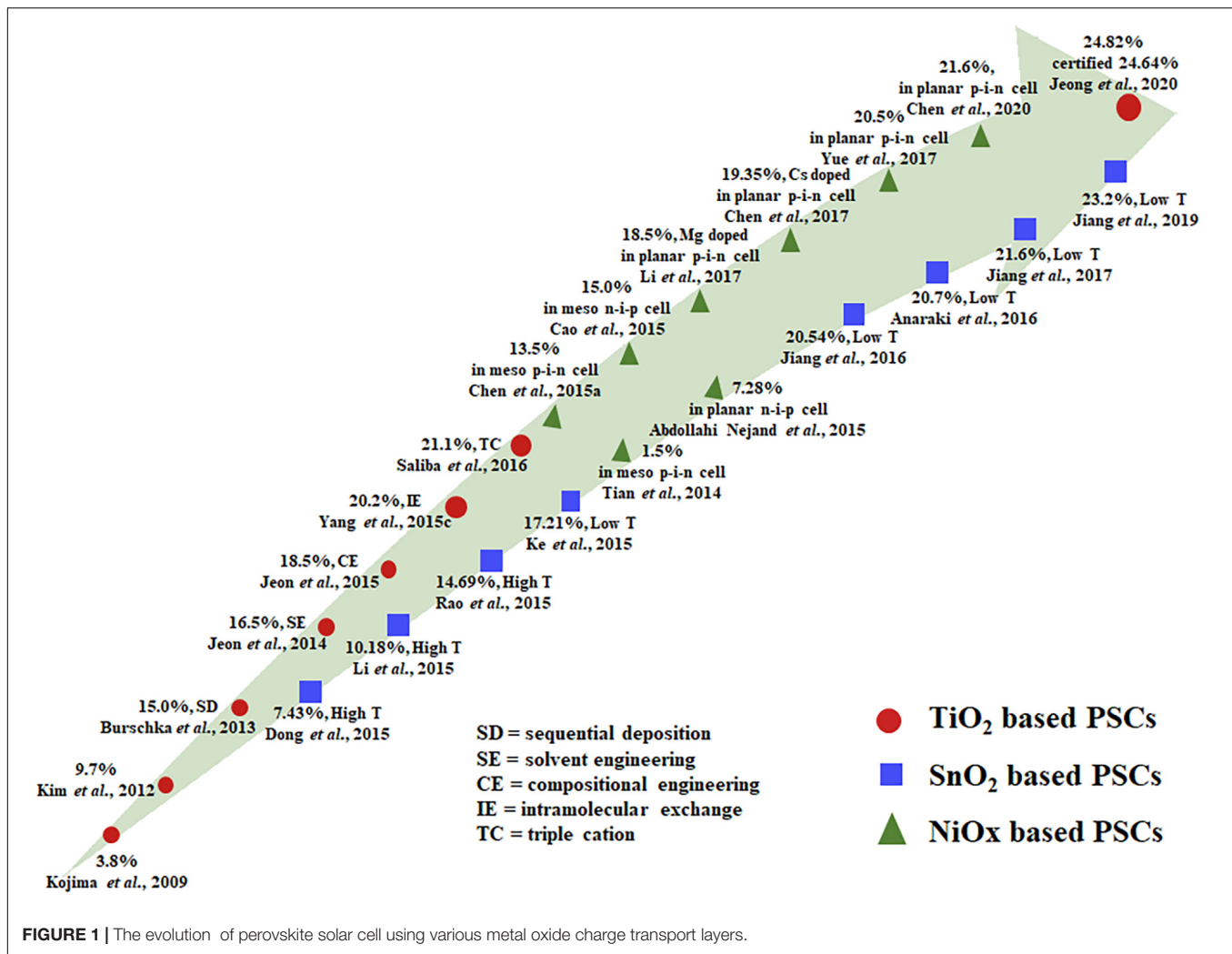


FIGURE 1 | The evolution of perovskite solar cell using various metal oxide charge transport layers.

have been commonly used as HTMs for PSCs. In 2014, Tian et al. (2014) firstly introduced the p-type NiO mesoporous for perovskite-sensitized solar cells, achieving a PCE of 1.5%. The group of Ahmadi demonstrated NiO<sub>x</sub>/Ni double layer as HTL deposited by the sputtering process in a planar n-i-p structure for PSCs, yielding a PCE of 7.28% (Abdollahi Nejad et al., 2015). In 2015, the group of Han employed a 10–20 nm NiO compact layer along with mesoporous Al<sub>2</sub>O<sub>3</sub> scaffold as HTM in p-i-n-based PSCs, obtaining a promising efficiency of 13.5% (Chen et al., 2015a). The group of Wang reported using NiO<sub>x</sub> HTM in mesoporous n-i-p-based PSCs with a further enhanced PCE to 15.03% (Cao et al., 2015). Li et al. (2017) applied sputtered NiO<sub>x</sub> for planar PSCs, which exhibit a champion PCE of 18.5%. Furthermore, introduction of dopants, commonly Li (Qiu et al., 2017), Co (Natu et al., 2012), Mg (Chen et al., 2015b), Cu (Chen et al., 2018), Cs (Singh et al., 2020), etc., in NiO<sub>x</sub> can further enhance the charge transport properties due to improved carrier conductivity as well as better energy level alignment across the interface of perovskite and HTL. The group of He and Lei doped NiO<sub>x</sub> with Cs and Cu, yielding a PCE of 19.35% (Chen et al., 2017) and 20.5% (Yue et al., 2017) for planar p-i-n-based PSCs, respectively. Recently, a latest high PCE of

21.6% was achieved for NiO<sub>x</sub>-based PSCs in a p-i-n structure after passivating the perovskite/ETL by using Cd<sub>x</sub>Zn<sub>1-x</sub>Se<sub>y</sub>S<sub>1-y</sub> quantum dots (Chen et al., 2020). The representative works for NiO<sub>x</sub>-based PSCs are summarized in Figure 1 and indicated in solid triangles.

## THE CRITERIA OF EFFECTIVE METAL OXIDE CTLS

The appropriate energy band alignment between CTLS and the perovskite absorbing layer is one of the fundamental criteria to ensure efficient charge extraction in PSCs. The conduction band minimum (CBM) of ETL is slightly lower than the conduction band of the perovskite, while the valence band maximum (VBM) of HTL is slightly higher than the valence band of the perovskite so that the CTLS can extract the corresponding charge carriers readily. Meanwhile, the energy barrier between the VBM of ETL and perovskite (and barrier between CBM of HTL and perovskite) is sufficiently large to block the counter carriers generated from the perovskite layer, suppressing the opportunity of charge recombination within the devices. The

built-in potential ( $V_{bi}$ ) can be influenced by the types of CTMs and the interfacial quality between the CTL and perovskite (Lin et al., 2017; Shin et al., 2019). The larger  $V_{bi}$  of devices enables PSCs to have a stronger ability to separate charges and transport and collect photo-generated carriers, yielding a higher  $V_{OC}$ . A variety of research works have demonstrated that minimizing the interfacial charge recombination between CTL and perovskite layer can effectively enhance the  $V_{OC}$  of PSCs (Jung et al., 2019; Kaneko et al., 2019; Aidarkhanov et al., 2020). Carrier mobility is another critical factor affecting the charge transport properties. Carrier mobility should be high to avoid charge accumulation at the interfaces of CTL/perovskite. The bandgap of CTMs should be wide and possess small refractive indexes so that the amount of visible light penetrating through the CTL to the perovskite absorber layer in the PSCs can be maximized.

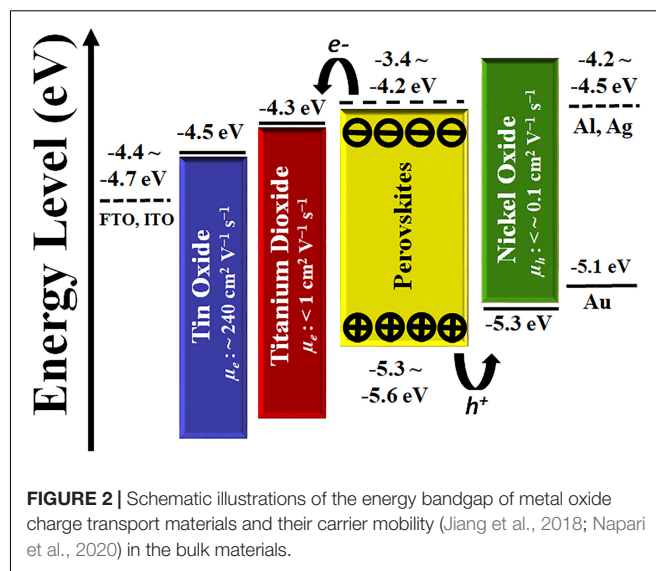
The morphology of CTLs should be controlled well in PSCs. The underlying ETL or HTL will determine the morphology of the perovskite film deposited on the top of CTLs for n-i-p or p-i-n device structures, respectively. It is known that the optimized morphology of the CTL-coated substrate can assist in the crystallization of the subsequent depositing perovskite layer and improve the perovskite film coverage on the CTL, inhibiting the formation of pinholes to prevent the introduction of undesired shunt paths in PSCs (Ng et al., 2016; Wang et al., 2019). Furthermore, the interfacial quality between the CTL and perovskite, which is of significant importance to determine the PCE and stability of PSCs, is strongly influenced by the surface morphology of CTLs. Intensive research works have exhibited different strategies to passivate the defects, which likely concentrate at material interfaces to suppress charge recombination and hysteresis effect (Shao and Loi, 2020). Meanwhile, the interface between the CTL and conductive electrode should also be optimized for forming of a good ohmic contact for PSCs (Babaei et al., 2020; Tseng et al., 2020).

It is noteworthy that metal oxide materials used as CTLs should be insensitive to ultraviolet light and possess very low photocatalytic effect in order to maintain a long-term stability of PSCs operating under strong sunlight. Metal oxide CTLs, unlike organic materials such as the commonly used PEDOT:PSS HTL, are usually not hygroscopic, which are more robust to moisture, allowing them to be the protection layers and preventing the perovskite active layer from degradation in the humid ambient air. Nowadays, metal oxide CTLs have shown their outstanding chemical stability, which is one of the essential factors for achieving long-lifetime PSCs for future commercialization (You et al., 2016; Lei et al., 2019; Singh et al., 2019; Thambidurai et al., 2020). **Figure 2** indicates the general properties of three different CTMs as discussed in this work.

## PREPARATION METHODS FOR METAL OXIDE CTLs

### Solution-Process Methods

The solution process is a commonly used method for depositing a wide range of metal oxides in thin films and different



nanostructures. This method is cost-effective and vacuum-free and allows low-temperature processing. In general, most of the metal oxide CTLs employed in PSCs are deposited by spin-coating processes. Other common solution techniques include sol-gel methods (Wojciechowski et al., 2014; Zhu et al., 2014; Ke et al., 2015), sono-chemical (Rashad et al., 2014), chemical precipitation (Ghosh et al., 2006), spray pyrolysis (Yue et al., 2017), chemical combustion (Jung et al., 2015), chemical bath deposition (Xia et al., 2008), etc. A number of comprehensive reviews on solution-processed metal oxide CTLs for application in solar cells have been published in the community (Jiang et al., 2017; Cao et al., 2018; Elseman et al., 2020b).

### Atomic Layer Deposition

Atomic layer deposition (ALD) is another method to prepare metal oxide CTLs with the film thickness precisely controlled at the atomic scale with excellent uniformity (Meng et al., 2011). Plenty of works have previously exhibited that ALD method is capable of forming high-quality metal oxides for PSC applications (Dong et al., 2014; Kim et al., 2016; Wang et al., 2016). The highest PCE of PSCs with metal oxide CTLs (using  $\text{SnO}_2$ ) formed by ALD was over 20%, along with a high  $V_{OC}$  of 1.23 V (Correa-Baena et al., 2017).

### Mechanosynthesis for Low-Dimensional Metal Oxides

Preparation of metal oxides by physical grinding methods has been used in organic solar cells and PSCs (Huang et al., 2012; Singh et al., 2018). The grinding process is clean, has low cost, and has high yield for mass production, which can produce high-level-purity and high-level-crystallinity metal oxide nanoparticles with controllable size and shape while keeping their intrinsic properties (Elseman et al., 2016; Singh et al., 2018). Grinding is a high-energy wet milling process where three-dimensional

**TABLE 1** | Classification of some common preparation methods for charge transport layers (CTLs) in terms of cost and processing temperatures.

Method	Temperature (°C)	Cost	Example of CTLs	References
Sol-gel	Low/high	High/low	TiO <sub>2</sub> , SnO <sub>2</sub> , NiO <sub>x</sub>	Wojciechowski et al., 2014; Zhu et al., 2014; Ke et al., 2015
Atomic layer deposition	Low	High	SnO <sub>2</sub> , TiO <sub>2</sub>	Tilley et al., 2014; Baena et al., 2015
Thermal decomposition	High (≥400)	High	NiO <sub>x</sub>	Zhang et al., 2008
Hydrothermal	Low	Low	NiO, TiO <sub>2</sub>	Sajid et al., 2019; Elseman et al., 2020c
Sono-chemical	Low	High	NiO <sub>x</sub>	Rashad et al., 2014
Chemical precipitation	Low	High	NiO <sub>x</sub>	Ghosh et al., 2006
Chemical combustion	Low	Low	Ni <sub>2</sub> O <sub>3</sub>	Elseman et al., 2020a
Chemical combustion	Low (≤150)	Low	NiO <sub>x</sub>	Jung et al., 2015
Thermal evaporation	High (≥400)	High	NiO <sub>x</sub> , TiO <sub>2</sub>	Khan et al., 2018; Kim et al., 2020
Electrodeposition	High (≥500)	Low	TiO <sub>2</sub>	Su et al., 2015
E-beam evaporation	Low	Low	SnO <sub>2</sub>	Ma et al., 2017
Pulsed laser deposition	High (≥400)	High	NiO <sub>x</sub>	Wang et al., 2012
Sputtering	Low	High	NiO <sub>x</sub> , TiO <sub>2</sub>	Wang et al., 2014; Yang et al., 2015b
Spray-pyrolysis	High (≥400)	Low	NiO <sub>x</sub>	Yue et al., 2017
Chemical bath deposition	Low (≤150)	Low	NiO, TiO <sub>2</sub> , SnO <sub>2</sub>	Xia et al., 2008; Dou et al., 2011; Wu et al., 2018
Ball-milling	Low (≤150)	Low	TiO <sub>2</sub> , SnO <sub>2</sub> , NiO	Singh et al., 2018; Singh et al., 2019; Singh et al., 2020

materials are broken down into lower-dimensional materials such as nanoparticles, nanorods, nanobelts, nanosheets, and nanofibers *via* controlling the milling conditions (solvents, temperature, and milling period) (Ibrahem et al., 2014; Ding et al., 2017). This method has been recently demonstrated for preparing TiO<sub>2</sub>, SnO<sub>2</sub>, and NiO<sub>x</sub> CTLs in PSCs (Singh et al., 2018, 2019, 2020). The as-prepared metal oxide nanomaterials can form a compact CTL in PSCs by low-temperature processing methods such as spin-coating the nanomaterial dispersion solution (Singh et al., 2018), which allows the fabrication of devices on flexible substrates or preparation of metal oxides on top of the perovskite layer without introduction of thermal degradation.

## Other Deposition Methods

There are other effective deposition methods to prepare the metal oxides for PSCs. Electrodeposition (ED) is one of cost-effective and scalable deposition processes with controllable film thickness. The group of Wei employed the technique of ED to prepare an ultrathin film of TiO<sub>2</sub> layer, yielding the PSC with a PCE of 13.6% (Su et al., 2015). In addition, other Physical vapor deposition (PVD) methods have been used to deposit the metal oxide CTLs such as magnetron sputtering (Yang et al., 2015a), pulsed layer deposition (Yang et al., 2016), and e-beam evaporation (Ma et al., 2017). The metal oxide CTL deposition route can be evaluated based on the cost and temperature. PVD and chemical vapor deposition (CVD) routes are regarded as high-cost methods due to their high-power consumption during the deposition process. There are a variety of deposition methods classified under CVD and PVD, such as thermal evaporation, pulsed laser deposition, DC and RF sputtering, plasma-enhanced CVD, metal-organic CVD, and low-pressure CVD. Compared to the techniques of PVD and CVD, chemical solution deposition is cheaper and more accessible. The

chemical solution deposition process is classified into different types such as chemical bath deposition, spin-coating, dip coating, screen printing, and spray pyrolysis. The common deposition routes for metal oxide CTLs are classified in terms of the cost and the required temperature for processing, which are shown in **Table 1**.

## PERSPECTIVE

The stability of PSCs can be enhanced by incorporating metal oxide CTLs for both ETL and HTL in the same device. However, nowadays, device stability remains a challenge since the majority of PSCs employ metal oxide CTL as either ETL or HTL while using organic or fullerene-based materials as the counter-CTL. For PSCs with all-inorganic CTLs, the processing temperatures of the metal oxide layer, which is located above the perovskite, usually should be below 100°C to prevent the thermal decomposition of the perovskite absorber. Spin-coating of a dispersion solution containing pre-synthesized metal oxide nanostructures on top of the perovskite is one of the possible techniques to yield metal oxide CTL at a low temperature. Considering the commercial standards for practical PSCs, the preparation methods for metal oxide CTLs should be cost-effective and compatible with low-processing temperature for flexible substrates as well as scalable for large solar modules. The mechanical flexibility of metal oxide CTLs should be also carefully investigated together with the perovskite absorber and flexible substrates during the blending process. Meanwhile, the interfacial engineering between the perovskite absorber and the CTL should be considered as well to modify the interface properties and passivate the defects, which is an important strategy to enhance device efficiency and stability.

## DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author/s.

## AUTHOR CONTRIBUTIONS

MS conceived the idea. MS and AN wrote the manuscript. AN and CC were involved in the manuscript discussion and

correction. All authors contributed to the article and approved the submitted version.

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- Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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