

Spray-coated SnO₂ electron transport layer with high uniformity for planar perovskite solar cells

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Abstract SnO₂ has been proven to be an effective electron transport layer (ETL) material for perovskite solar cells (PSCs) owing to its excellent electrical and optical properties. Here, we introduce a viable spray coating method for the preparation of SnO₂ films. Then, we employ a SnO₂ film prepared using the spray coating method as an ETL for PSCs. The PSC based on the spray-coated SnO₂ ETL achieves a power conversion efficiency of 17.78%, which is comparable to that of PSCs based on conventional spin-coated SnO₂ films. The large-area SnO₂ films prepared by spray coating exhibit good repeatability for device performance. This study shows that SnO₂ films prepared by spray coating can be applied as ETLs for stable and high-efficiency PSCs. Because the proposed method involves low material consumption, it enables the low-cost and large-scale production of PSCs.

Keywords spray coating, SnO₂ film, ETL, PSCs

1 Introduction

Organic–inorganic hybrid perovskite solar cells (PSCs) have received much attention in recent years owing to their low manufacturing cost and excellent photovoltaic performance [1]. The maximum power conversion efficiency (PCE) has been increased from 3.8% in 2009 to 24.2% in 2019, showing great potential for PSCs to replace traditional silicon solar cells [2–5]. Because the PCE is close to the theoretical efficiency limit, it is very important to develop fabrication techniques for high durability, low cost, and flexible devices, for which the low-temperature preparation of electron transport layers (ETLs) is a key

technology [6,7]. Current high-efficiency PSCs typically use TiO₂ as the ETL. However, devices based on TiO₂ ETLs typically require TiO₂ to be annealed at temperatures above 450°C, which is detrimental to future commercial applications [8–11]. Other low-temperature processed materials, such as SnO₂ [12–15], WO₃ [16], ZnO [17,18], amorphous-TiO_x [19], and [6,6]-Phenyl-C61-butyric acid methyl ester [20,21], have been studied as ETLs to replace TiO₂. Despite its low-temperature processability, SnO₂ is regarded as the most promising alternative to TiO₂ owing to its deep conduction band, high electron mobility, high light transmittance, good ultraviolet resistance, and excellent chemical stability. SnO₂-based planar PSCs yield a high PCE close to 21%, which is comparable to that of TiO₂-based planar PSCs.

In order to obtain SnO₂-based ETLs, various methods have been studied, including spin coating and hydrothermal and chemical bath depositions [22–26]. However, these methods have limitations. For instance, only small- or large-area films with poor optical properties can be obtained, and it is difficult to obtain large-area SnO₂ films with excellent performance. Moreover, these methods usually require cumbersome production processes and produce a large amount of material waste, which is not conducive to low-cost and large-scale commercial production [27–30]. Some studies have reported the preparation of PSCs based on spray-coated TiO₂, but with low efficiencies comparable to those of solar cells based on films prepared by spin coating [11]. There are also some studies on the preparation of SnO₂ films by spraying SnCl₂ precursor, but the optical properties of these SnO₂ films are not good enough [31,32]. To solve this, we developed a spray coating method to prepare SnO₂ films using SnO₂ hydrocolloid instead of SnCl₂ precursor, which enables a more straightforward and efficient preparation of large-area SnO₂ films with excellent optical performance. In particular, the morphologies of SnO₂ films can be

conveniently controlled by changing the spraying conditions, such as the substrate temperature, spraying time, solution flow rate, and nozzle-to-substrate distance. This method can also be used to improve the material utilization and, thus, reduce the manufacturing cost of PSCs [33–37].

In this work, we report a universal low-cost spray coating process for the preparation of SnO₂ films in an air environment and use it to prepare an ETL for PSCs. Specifically, dense and smooth SnO₂ films are produced by optimizing the spray conditions and film thickness to obtain the best PSCs. A dozen of devices fabricated from the same batch of spray-coated SnO₂ films showed similar high performance with an average efficiency of over 16%, demonstrating the reliability of the large-area SnO₂ films prepared by spray coating. The PSC based on a spray-coated SnO₂ film achieved a maximum conversion efficiency of 17.78%, which is almost the same as that of PSCs based on SnO₂ prepared by spin coating. Furthermore, the device based on the spray-coated SnO₂ ETL showed better device stability than the PSC based on SnO₂ prepared by spin coating.

2 Experimental

2.1 Materials

All chemicals were applied to the experiments without further purification, and all solvents were of super dehydrated grade. SnO₂, 15 wt-% in H₂O colloidal dispersion was purchased from Alfa Aesar. All perovskite materials (including HC(NH₂)₂I (≥99.5%), CH₃NH₃Br (≥99.5%), PbI₂ (≥99.99%), PbBr₂ (≥99.99%), and CsI (≥99.99%)), 2,2',7,7'-tetrakis-(*N,N*-di-*p*-methoxyphenylamino)-9,9'-spirobifluorene (Spiro-OMeTAD, ≥99%) and lithium-bis(tri-fluoromethanesulfonyl)imide (≥99%) were purchased from Xi'an Polymer Light Technology Corp. 4-*tert*-Butyl pyridine (96%) and chlorobenzene (99.8%) were purchased from Sigma Aldrich. *N,N*-Dimethylformamide (99.7%) and dimethyl sulfoxide (99.8%) were purchased from Beijing Infinity Scientific.

2.2 Preparation of perovskite solar cells

The per-patterned indium tin oxide (ITO) glass substrates were cleaned with detergent, deionized water, acetone and isopropanol in an ultrasonic bath for 15 min, respectively. Before spin coating or spray coating, the substrates were treated with ultraviolet ozone cleaner for 15 min. SnO₂ in H₂O colloidal dispersion was diluted to different concentrations with ultrapure water and stirred for 30 min. A schematic view of the spray coating device is shown in Fig. S1(a) (cf. Electronic Supplementary Material, ESM). The spray nozzle was placed 20 cm above the surface of the substrate and the diluted SnO₂ colloidal dispersion was sprayed at a flow rate of 0.2 mL·min⁻¹ for different

duration. SnO₂ hydrocolloid (1 wt-%) was sprayed on ITO substrates for different duration to get various film thicknesses and then annealed at 150°C for 30 min. SnO₂ hydrocolloid (3 wt-%) was spin-coated on ITO substrates at 3000 r·min⁻¹ for 30 s and then annealed at 150°C for 30 min. Then, the precursor solution of Cs_{0.05}FA_{0.85}MA_{0.15}PbI_{2.9}Br_{0.15} was spin-coated at 5000 r·min⁻¹ for 35 s in a nitrogen glove box, and 300 μL chlorobenzene was dropped 10 s after the beginning of spin-coating. And the substrates were annealed at 100°C for 60 min. After that, 80 mg Spiro-OMeTAD was dissolved in 1 mL chlorobenzene with additive of 4-*tert*-butylpyridine (10.5 μL) and 0.2 mol·L⁻¹ solution of lithium-bis(tri-fluoromethanesulfonyl)imide in acetonitrile (15.5 μL). The Spiro-OMeTAD solution was spin-coated on perovskite film at 4000 r·min⁻¹ for 30 s and place for more than 6 h in dry air. Finally, 60 nm-thick silver layer was vacuum-evaporated as the counter electrode to give a complete device.

2.3 Characterization

The morphologies of SnO₂ films and perovskite films were observed using scanning electron microscopy (SEM, Hitachi SU8000). The surface morphologies of SnO₂ films were analyzed through atomic force microscopy (AFM) in tapping mode under ambient conditions using a Bruker instrument. The thickness of SnO₂ films were analyzed using Dektak XT. The X-ray diffraction (XRD) patterns were recorded on Bruker D8 X-ray diffractometer with CuKα radiation (λ = 1.5418 Å) at 25°C. The data were collected with a 0.02° step size (2θ) for 0.2 s. The current density-voltage (*J-V*) characteristics of solar cells were measured by a computer-controlled source meter (Keithley 2400) under simulated one sun illumination (AM 1.5G, 100 mW·cm⁻²), as calibrated by a reference monocrystalline silicon solar cell (91150 V Oriel Instruments). The active area of the cell was fixed to be 0.04 cm² with a non-reflective metal mask. The external quantum efficiency (EQE) spectra were measured in air under short-circuit conditions using Crowntech QTest Station 1000AD equipped with a 100 W Xe arc lamp, a filter wheel, and a monochromator.

3 Results and discussion

To prepare SnO₂ ETLs by spray coating for PSCs, we firstly determined some spray parameters, including the concentration of SnO₂ in H₂O colloidal dispersion, the flow rate of the gun, and the spraying time, which play an important role in the morphology of the prepared SnO₂ film. It has been found that the thickness and compactness of SnO₂ films are very important for high performance. Therefore, we chose a SnO₂ hydrocolloid with a lower concentration (1 wt-%) for spray coating than that for spin

coating (3 wt-%), which will make it easier to adjust the thickness and surface morphology of the SnO₂ films. Through preliminary tests, we found that the SnO₂ films become too thick and uneven, and their transmittance is deteriorated after being sprayed for more than one minute. The thickness of an SnO₂ film prepared by spraying for one minute was approximately 100–110 nm, which is too large for application in PSCs. When sprayed for 20 s, a smooth, dense, and high light-transmitting SnO₂ film with a thickness of ~30–35 nm, which is suitable for application in PSCs, was obtained. The thickness of SnO₂ films at different spraying times is shown in Fig. S2. Below, the surface morphology of a SnO₂ film spray coated at a flow rate of 0.2 mL · min⁻¹ for 20 s will be compared with that of traditional spin-coated SnO₂ films.

Because SnO₂ droplets sprayed from a spray gun are not completely uniform, when they are sprayed onto ITO substrate, there is no guarantee that their density distribution on the surface will be uniform. This issue can be mitigated by spraying for a long time. However, unlike with spin coating, the volatilization process of SnO₂ droplets on the ITO substrate will affect the surface morphology of the resulting SnO₂ films [27,34]. To study the difference in surface morphology of SnO₂ films prepared by spray coating and spin coating, we first investigate their SEM images. Figures 1(a) and 1(b) show the top-view SEM images of SnO₂ films prepared by spray coating for 20 s and spin coating, respectively. It can be seen that both films are flat and dense, and there are no holes on their surface.

Although the SEM images prove that the SnO₂ film prepared by spray coating is dense, its surface roughness cannot be evaluated from these images. Accordingly, we collected AFM images of the SnO₂ films prepared by the two methods. Figures 2(a,b) and 2(c,d) show the AFM images of the SnO₂ films prepared by spray coating for 20 s and spin coating, respectively. Figures 2(a) and 2(c) show that, as expected, the SnO₂ film prepared by spray coating is rougher than that prepared by spin coating. The root-mean-square roughness (RMS) value of the SnO₂ film prepared by spray coating is 2.24 nm, which is almost twice that of the spin-coated film (RMS = 1.11 nm). Nevertheless, the surface roughness is still very small

relative to the perovskite film thickness of several hundred nanometers typically used in PSCs, and this degree of roughness does not affect the perovskite layer.

To determine the performance of the SnO₂ film prepared by spray coating, we fabricate devices with the structure of ITO/SnO₂/Cs_{0.05}FA_{0.85}MA_{0.15}PbI_{2.9}Br_{0.15}/Spiro-OMe-TAD/Ag. However, the formation of perovskite may be affected by the different surface morphology of the SnO₂ films prepared by spray coating and spin coating. To understand this impact, we performed XRD and SEM analysis of the perovskite films prepared with the different SnO₂ films, as shown in Figs. 3 and S3, respectively. No difference between the two perovskite films was observed, so we believe that the slight difference in surface morphology of the SnO₂ films does not affect the formation of perovskite. Considering that different spray coating times will lead to SnO₂ films with different thicknesses, we tested different spray coating times from 10 to 50 s to determine the performance of the SnO₂ films. The corresponding photovoltaic parameters of the PSCs are summarized in Fig. 4 and Table 1. It can be seen that the performance of the PSC based on the SnO₂ film prepared by spraying for 10 s is poor. This is because the SnO₂ film cannot fully cover the ITO surface, which forms a shunting path. With the spraying time of 20 s, both short-circuit current density (J_{sc}) and fill factor (FF) are improved, reaching maximum values of 23.35 mA · cm⁻² and 0.666, respectively. As a result, the device achieved a maximum PCE of 17.78%. Such improvement may be attributed to the changes in the SnO₂ film thickness. An appropriate thickness of the SnO₂ film facilitates electron extraction. With longer spraying times, as the spraying time increase, J_{sc} and FF decrease. This is because when the spraying time is too long, the SnO₂ film becomes too thick, resulting in an increase in the series resistance (R_s) of the SnO₂ film.

To verify the effectiveness of the spray-coated SnO₂ ETL in PSCs, we compared the best device based on an SnO₂ ETL prepared by spray coating with that prepared by conventional spin coating, and the results are shown in Fig. 5 and Table 2. The two devices show very similar PCE values, J_{sc} , voltage (V_{oc}), FF , and R_s . The EQE response of the device based on the spray-coated SnO₂ ETL is slightly

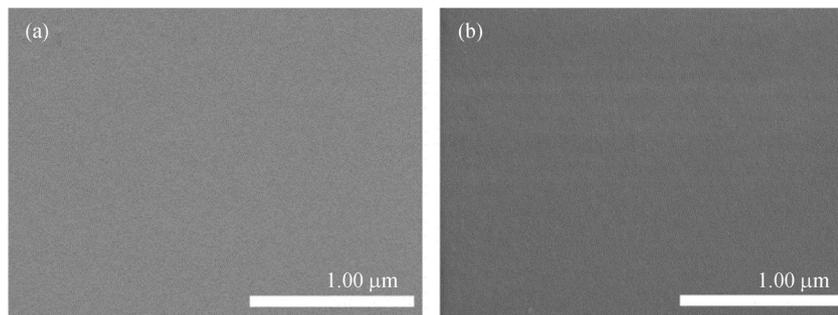


Fig. 1 SEM images of SnO₂ films prepared by (a) spray coating and (b) spin coating.

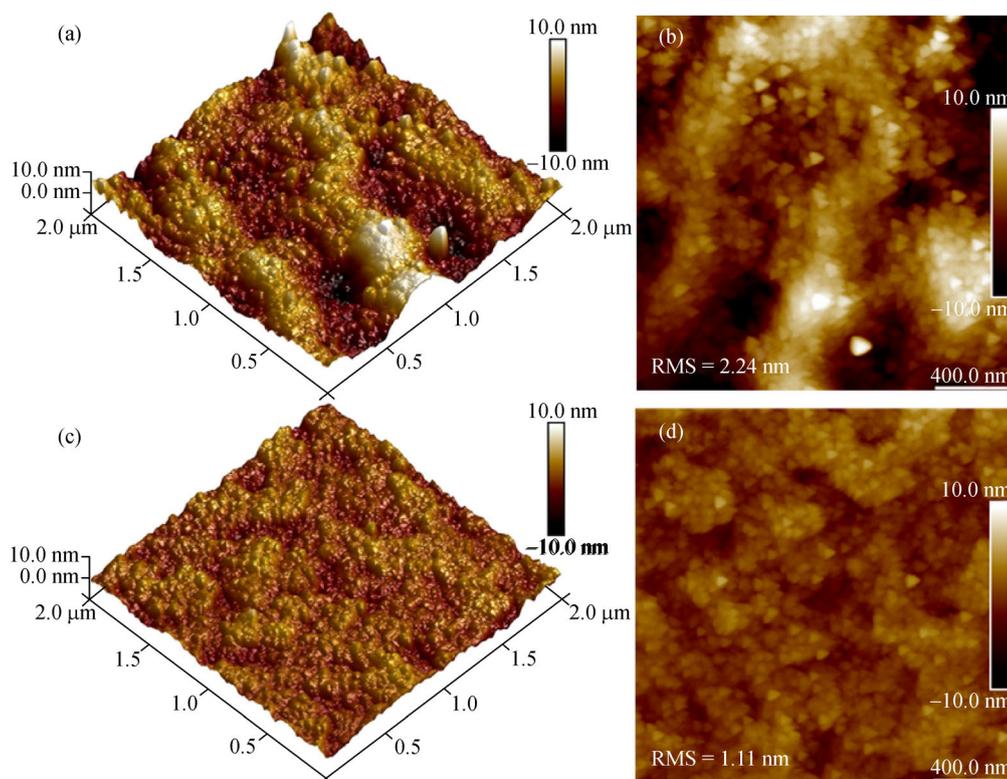


Fig. 2 AFM images of SnO₂ films prepared by (a,b) spray coating and (c,d) spin coating.

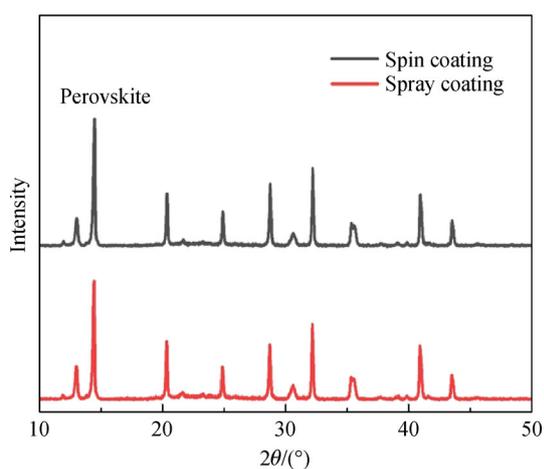


Fig. 3 XRD patterns of perovskite films deposited on SnO₂ films prepared by different methods.

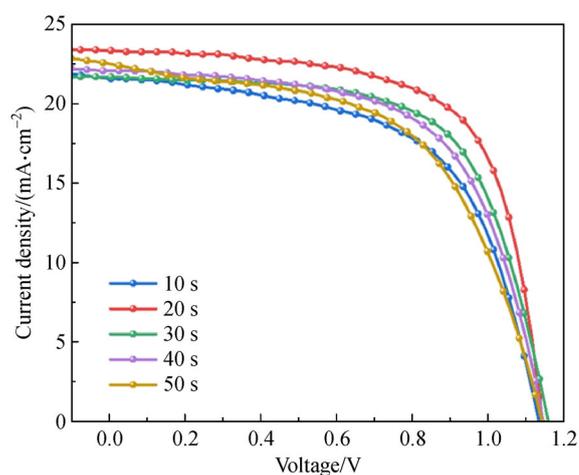


Fig. 4 J - V curves of PSCs based on SnO₂ prepared at different spraying times.

Table 1 Photovoltaic parameters of devices based on SnO₂ prepared at different spraying times

Spraying time/s	$J_{sc}/(\text{mA} \cdot \text{cm}^{-2})$	V_{oc}/V	FF	$PCE/\%$	R_s/Ω
10	21.61 ± 0.35	1.134 ± 0.009	0.592 ± 0.025	15.42 ± 0.50	240
20	23.35 ± 0.50	1.144 ± 0.010	0.666 ± 0.030	17.78 ± 0.42	156
30	21.69 ± 0.45	1.159 ± 0.011	0.642 ± 0.028	16.14 ± 0.39	196
40	22.03 ± 0.48	1.145 ± 0.011	0.616 ± 0.035	15.73 ± 0.56	233
50	22.51 ± 0.45	1.143 ± 0.010	0.559 ± 0.040	14.37 ± 0.58	266

higher than that of the device based on the spin-coated SnO₂ ETL. The integrated photocurrent of the SnO₂ ETL deposited by spin coating and spray coating, which were calculated from the EQE spectra, are 21.5 and 21.9 mA·cm⁻², respectively. This is consistent with the J_{sc} values of the J - V curves. These results show that the SnO₂ film prepared by spray coating can be used as an ETL for PSCs with performance similar to that of PSCs based on spin-coated SnO₂ films. In addition, we investigated the stability of the devices based on the spray- and spin-coated SnO₂ under simulated one-sun illumination in air without encapsulation (Fig. 6). The two devices show similar device stability. The PSC based on the spin-coated SnO₂ film, retained an efficiency of 85% of its initial value after storage for 600 h.

To confirm the performance of the large-area SnO₂ film

prepared by spray coating, we spliced together twelve pieces of ITO substrates with dimensions of 1.5 cm × 2 cm and sprayed them with SnO₂. Then, we used these twelve substrates to prepare PSCs under the same conditions. Figure 7 shows the PCEs of the twelve cells based on the same batch of spray-coated SnO₂ films. As shown in the figure, all the devices have high efficiency. The corresponding photovoltaic parameters of the PSCs are summarized in Table S1. The devices in the middle zone have the best efficiency of approximately 17.6%, and the average efficiency of all the devices is 16.45%. This confirms that the large-area SnO₂ film prepared by spray coating is very reliable, and the whole film is dense and uniform. It is worth mentioning that the SnO₂ film preparation by spraying does not require a specific environment, and can be directly prepared in air.

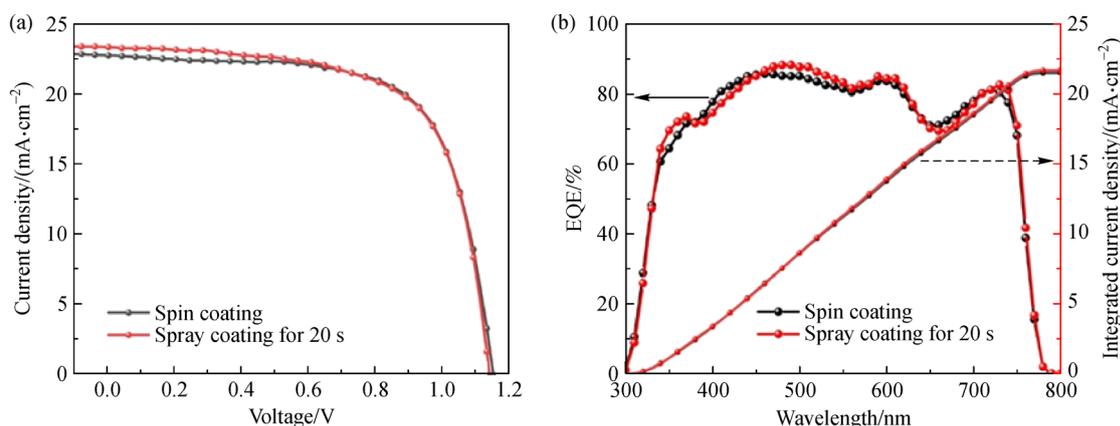


Fig. 5 (a) J - V curves and (b) EQE of PSCs based on SnO₂ prepared by spin coating and spray coating.

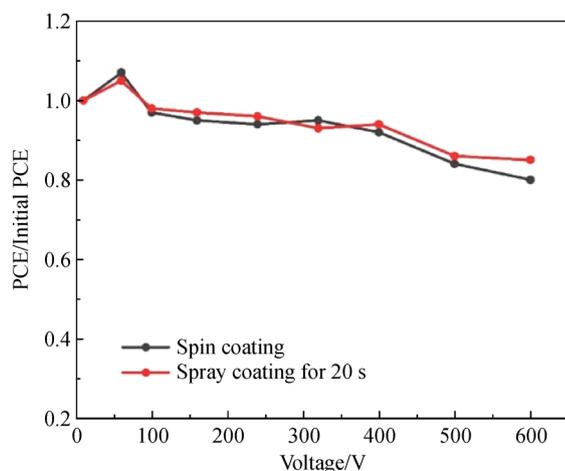


Fig. 6 Stability test of devices without any encapsulation.

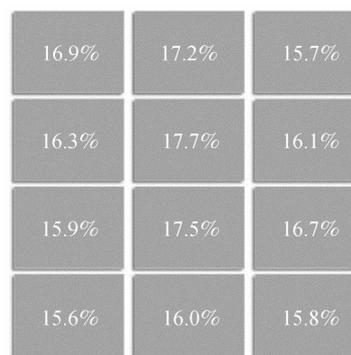


Fig. 7 PCEs of twelve PSCs based on the same batch of spray-coated SnO₂ films under the same conditions.

Table 2 Photovoltaic parameters of PSCs based on SnO₂ prepared by spin coating and spray coating for 20 s

Item	$J_{sc}/(\text{mA} \cdot \text{cm}^{-2})$	V_{oc}/V	FF	$PCE/\%$	R_s/Ω
Spin coating	22.80 ± 0.48	1.156 ± 0.010	0.679 ± 0.030	17.90 ± 0.39	160
Spray coating	23.35 ± 0.50	1.144 ± 0.010	0.666 ± 0.030	17.78 ± 0.42	156

Furthermore, almost all the way SnO₂ is prepared into the SnO₂ films with very little waste. These results indicate that the spray-coated large-area SnO₂ film has the characteristics of low cost and high output. This is clearly one of the most promising technologies for the commercial preparation of ETLs for PSCs.

4 Conclusions

In conclusion, we successfully developed a simple and reproducible spray method to prepare uniform and dense SnO₂ ETLs for PSCs. Parameters such as the precursor concentration, flow rate of the gun, and spray time for the formation of the SnO₂ films were evaluated. The optimal device based on a spray-coated SnO₂ ETL achieved an efficiency of 17.78%, showing the great potential of spray-coated SnO₂ film as alternative ETLs for highly efficient and stable PSCs. Besides, compared with the conventional spin coating method, the SnO₂ spray coating method has lower material consumption, larger producible area, and easier process. This is a promising way to reduce the manufacturing cost of PSCs and provides an inexpensive and efficient method for preparing large-area flexible PSCs.

Acknowledgements The authors gratefully acknowledge financial support from the National Natural Science Foundation of China (Grants No. 11974129) to Xiao-Feng Wang and the Fundamental Research Funds for the Central Universities, Jilin University.

Electronic Supplementary Material Supplementary material is available in the online version of this article at <https://doi.org/10.1007/s11705-020-1917-x> and is accessible for authorized users.

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