

REVIEW

Inorganic top electron transport layer for high performance inverted perovskite solar cells

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Funding information

Collaboration Research Fund, Grant/Award Number: C7035-20G; Collaboration Research Fund from Guangdong-Hong Kong-Macao Joint Laboratory, Grant/Award Number: 2019B121205001; General Research Fund from the research Grants Council of Hong Kong Special Administrative Region, Grant/Award Numbers: 17204117, 17200518, 17201819, 17211220; Major Basic Research Project of the Natural Science Foundation of the Jiangsu Higher Education Institutions, Grant/Award Number: 20KJB430023; Qinglan Project of Jiangsu Province of China, Grant/Award Number: 2021-11; The University Grant Council of the University of Hong Kong for the Funding, Grant/Award Numbers: 201811159147, 2019157209

Abstract

As promising photovoltaic devices, perovskite solar cells (PSCs) have attracted extensive and ongoing attention due to easy manufacturing and high power conversion efficiency (PCE). Although the PCE is lower than that of PSCs with normal structure, inverted PSCs have been widely investigated due to their lower hysteresis and potential application. Electron transport layer (ETL) on top of perovskite film in inverted PSCs plays a significant role in device performance. Inorganic top ETL has been used to replace organic ETL for their excellent characters. This review summarizes the progress of inorganic top ETL for high-performance inverted PSCs. Firstly, the principles of top ETL and advantages of inorganic ETL are highlighted. Then the established top ETLs are summarized. Subsequently, various strategies for top ETL fabrication are shown to demonstrate their advantages and shortcomings. Finally, conclusion and outlook of top ETL in inverted PSCs are presented, addressing the issues and directing the hopeful solutions.

KEYWORDS

high performance, inorganic, perovskite solar cell, top electron transport layer

1 | INTRODUCTION

In recent years, perovskite solar cells (PSCs) have gained worldwide attention from the lab to industry fabrication and PSC with a power conversion efficiency (PCE)

greater than 25% has been reported.¹ Compared with the champion PCE of normal PSCs, this value of inverted PSCs is certified as 22.3%.² Although the PCE value is some lower, inverted PSCs have some special advantages, such as much lower hysteresis and promising application

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on tandem solar cells, which makes them a famous star among the new-generation photovoltaic devices.^{3–5} As an important functional layer in high performance inverted PSC, electron transport layer on top of perovskite film (namely top ETL) play an irreplaceable role. Known to be a star ETL, fullerene and its derivative such as [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) has been widely used in inverted PSCs, exhibiting negligible hysteresis.^{6–10} However, difference between the Fermi level of PCBM and the work function of Ag electrode often results in Schottky barrier at their interface,¹¹ leading to charge carrier recombination and high series resistance (R_s) and thus low PSC performance. To address these issues, inorganic top ETLs have been used due to their advantages such as good optical and electrical properties, low cost, facile synthesis, controllable properties, and so forth.

In this review, we first summarize the principles of top ETLs and the advantages of inorganic materials as top ETLs. Then the reported top ETLs and their fabrication/modification methods are presented. Finally, the conclusion and outlook are shown, containing some potential alternatives as inorganic top ETL for inverted PSCs.

2 | TOP ETL FOR INVERTED PEROVSKITE SOLAR CELLS

Planar PSCs (shown in Figure 1) are generally divided into n-i-p (regular) and p-i-n (inverted) types, which are respectively with the structure of transparent conductive oxide (ITO or FTO)/ETL/perovskite/HTL/rear electrode (Ag, Al, or Au) and transparent conductive oxide (ITO or FTO)/HTL/perovskite/ETL/rear electrode (Ag, Al, or Au). The PSCs work as the following steps: (a) the active layer (perovskite) absorbs photons under sun illumination; (b) photo-generated carriers emerge and separate into holes and electrons; (c) holes and electrons respectively transport to electrodes through HTL and ETL; (d) current engenders in the external circuit. Inverted PSCs have been widely studied for nearly 10 years and the first one with a low efficiency value of 3.9% was proposed in 2013 with the structure of ITO/poly

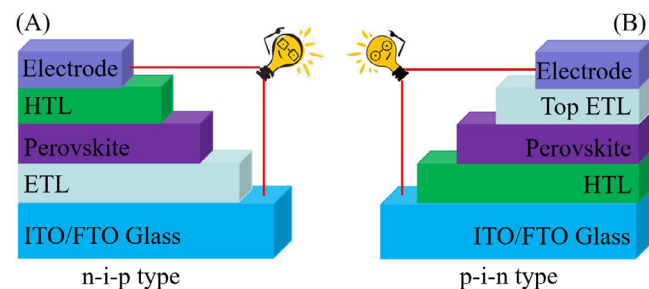


FIGURE 1 Structure of (A) n-i-p type PSC and (B) p-i-n type PSC

(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/CH₃NH₃PbI₃(MAPbI₃)/1-(2-methoxycarbonyl)-propyl-1-phenyl-(6,6)-C₆₁ (PCBM)/bathocuproine (BCP)/aluminum (Al).¹² Up to present, the champion PCE of inverted PSC is 22.3% (certified)¹³ for a small area and 18% for large area of 19.276 cm².¹⁴ Meanwhile, a mini-module of 36 cm² area with p-i-n structure first passed the 85°C/85% RH test according to the standards of IEC 61215 in 2019,¹⁵ indicating that inverted PSCs may be more suitable for commercialization.

2.1 | Principles of top ETL

Through the development of inverted PSCs, it is known to us that ETL atop the perovskite layer plays critical role both in device PCE and stability. The principles of top ETL in inverted PSCs are shown in Figure 2 and described as follows:

1. **Facile fabrication**, such as solution process, low-temperature even no annealing, and so forth. One advantage of PSCs is solution process.¹⁶ To lower the fabrication difficulty of PSCs, solution process for ETL is needed. Additionally, annealing is used to remove the solvent left after the above solution process. However, high annealing temperature will destroy the underneath perovskite layer. Therefore, low-temperature annealing or no annealing can prevent the destruction of perovskite from high temperature, thus pledging high device performance.
2. **Suitable energy level**: Mismatching energy level in PSCs will result in non-radiative recombination at the interfaces and inefficient charge transport from perovskite to ETL and rear electrode, causing thermal potential loss and mainly reducing the open circuit voltage (V_{OC}) and fill factor (FF) of the devices.^{17–21} So, suitable energy level of ETL, both the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO), is critical to device performance. High quality top ETL with suitable energy level is beneficial to electron transporting and hole blocking.
3. **High electrical conductivity**: Electron transporting in both n-i-p and p-i-n PSCs affects the device performance. Higher electrical conductivity leads smaller R_s



FIGURE 2 Schematic diagram of the principles of top ETL

of PSC, which brings in higher short-circuit current (J_{SC}), higher FF, and lower hysteresis thus higher device performance.^{12–26}

4. **Interfaces:** From the working mechanism of inverted PSCs, we know that the electron–hole pairs are generated under sunlight soaking and separated into free carriers at the interface.¹⁶ Generally, there are two ETL-based interfaces in inverted PSCs, concluding interfaces of perovskite/ETL and ETL/rear electrode. Interfacial defects from the interface of perovskite/ETL will improve the possibility of non-radiative recombination, which lowers the device performance.^{27–30} Meanwhile, the difference of work function (WF) between the ETL and rear electrode would form a Schottky barrier at the interface of ETL/rear electrode, leading to inefficient electron transport and lower device performance.^{31–35} Additionally, interface modification can improve device performance through a better energy alignment, reduced recombination, trap passivation, excellent contact with other functional layers, and lower hysteresis.^{36–40} The interfaces affect the performance of inverted PSCs and interface engineering is an efficient strategy for performance improvement.
5. **Morphology:** The morphology of top ETL also influences the device performance. Excellent morphology of top ETL will result in good contact between perovskite and ETL, which increases electron extraction and transport and inhibits recombination.^{28,41–48} ETL on top of perovskite layer has another function to avert the contact of perovskite and rear electrode. The penetration of perovskite through ETL to metal electrode degrades the electrode and decrease the device performance.^{49,50} Homogeneous ETL can efficiently prevent direct contact between the perovskite and rear electrode and greatly reduce device shunting and current leakage, enhancing device performance.^{28,42,51–65} In addition, good morphology of ETL, such as smooth and full coverage, is beneficial for the deposition of rear electrode and facilitates the formation of ohmic contact between the ETL and rear electrode.^{66–71}
6. **Stability:** PSCs performance will decrease along with the operational time due to water, oxygen, light, and heat. As known to us, efficient encapsulating can resist water and oxygen and thus ensure the corresponding stability. Photo- and thermal-stability of inverted PSCs are mainly affected by the functional layers of charge transport layers (CTLs, concluding ETL, and hole transport layer [HTL]),^{72–91} perovskite layer,^{92–117} and rear electrode.^{16,118–142} Top ETL is an irreplaceable layer for high performance inverted PSC and inevitably resolve the device stability.^{3,6,29,34,42,45,63,65,71,81,143–175} Therefore, high stability of top ETL will enhance the stability of PSCs and promote the device commercialization.

7. **Low cost.** Known to all, one of the issues that impede the commercialization of PSCs is fabrication cost. Top ETL is responsible for a great part of the whole cost of inverted PSCs. High cost of fullerene derivatives, such as PCBM and its modification, is an obstacle to commercializing inverted PSCs.^{25,145,176–181} Low cost of top ETL fabrication is beneficial for the PSC commercialization.^{30,34,180,182–193}

2.2 | Advantages of inorganic top ETL

Organic materials have been widely used as top ETL in efficient inverted PSCs.²⁵ The most popular organic top ETLs are fullerenes (C_{60} and C_{70}) and their soluble form phenyl-C61-butyric acid methyl ester (PCBM) due to their low-temperature deposition process and efficient electron transporting.^{25,34,51,194–199} Among them, PCBM was used as ETL in most p-i-n PSC cases.^{7–10,200,201} However, there are some shortcoming for fullerene different electron transport materials (ETMs), such as narrow range of LUMO level, relatively poor morphological stability, and high cost of synthesis and purification.¹⁹⁹

As shown above, some intrinsic weak points exactly exist in organic top ETL. Many researchers have continuously worked on finding inorganic alternatives to boost higher PCE and stability with lower cost. Compared with organic top ETL, inorganic top ETL has the advantages of (a) facile fabrication process,^{50,64,168,202–205} (b) high intrinsic stability,^{3,4,18,32,50,206–209} (c) easily adjustable optical and electronic properties,^{3,28,34,35,42,153,159,208} (d) low-temperature or no post-treatment,^{26,41,50,155,168,190,203} (e) low cost,^{68,163,187,210,211} (f) additional function as passivate layer.^{7,36,175} All the advantages of inorganic top ETL accelerate its application in high-performance inverted PSCs.

To verify the excellent properties of inorganic top ETL, some characterizations should be done. Transmittance, absorption spectra, mobility, conductivity et al. are used to demonstrate the optical and electronic properties. Surface defect passivation due to top ETL can be analyzed through the change of defect density. Many other characterizations not mentioned here are also used to illustrate the improved properties of inorganic ETL.

2.3 | Established inorganic top ETL

Inorganic top ETL for inverted PSCs has gained wide attentions due to above advantages. Metal oxides and their modified derivatives (such as doping with other elements and mixed with other materials)

TABLE 1 Photovoltaic parameters of PSCs based on inorganic top ETL

	Architecture	PCE (%)	Stability	Hysteresis (%)	Reference
Organic-inorganic multilayer ETLs	FTO/PEDOT:PSS/PVK/PCBM/TiO _x /Al	9.8	NM	NM	194
	ITO/GO/PVK/PCBM/ZnO/Al	12.40	NM	NM	20
	ITO/PEDOT:PSS/PVK/PCBM/Polymer/ZnO/Al	8.15	NM	NM	212
	FTO/NiO _x /PVK/PCBM/TiO _x /Ag	20.65	① > 85% after 500 h, continuous light illumination ② > 980% after 500 h, at 85°C	NM	144
	ITO/PEDOT:PSS/PVK/rGO:PCBM/PEN/Ag	14.5	>50% after 120 h, continuous light soaking at ~50% RH, unencapsulated	Free	27
	ITO/PEDOT:PSS/PVK/di-PDI + DMBI/TiO ₂ /Al	10.0	NM	NM	218
	FTO/NiMgLiO _x /PVK/PCBM/Ti(Nb)O _x /Ag	20.65	① > 85% after 500 h, continuous light soaking (429 nm UV cut off) under ambient condition, encapsulated ② > 90% after 500 h, encapsulated in dark at 85°C	NM	144
	ITO/P3CT-K/CuZnS/PVK/PCBM/ZnO/Al	18.30	89% after 72 h	3.83	149
	ITO/PTAA/PVK/C ₆₀ /ZnSe/Ag	18.38	NM	NM	224
	ITO/PEDOT:PSS/PVK/Br-PDI/ZnO NP/Ag	10.50	NM	NM	214
	ITO/NiO/PVK/PCBM/BCP/Cr/CrO ₃ /Au	18.5	After 1000 h, AM 1.5G, at 70–75°C	NM	156
	ITO/CuI/PEDOT:PSS/PVK/PC ₆₁ BM/ZnO/Al	14.3	88% after 720 h, unencapsulated in glovebox	NM	150
	FTO/Cu@CuI/PVK/PCBM/ZnO/Ag	18.8	90% after 1000 h, at RT with ~25% RH	NM	157
	ITO/CuI/PEDOT:PSS/PVK/PC ₆₁ BM/ZnO/Al	14.3	88% after 720 h, nonencapsulated in glovebox	NM	150
	ITO/Spiro-TTB/PVK/LiF/C ₆₀ /ALD-SnO ₂ /IZO/Ag	~11.3	90% after 1000 h, dark at 85°C with 85% RH	NM	151
	ITO/NiO/PVK/LiF/PC ₆₀ BM/SnO ₂ /ZTO/ITO/LiF/Ag	12.5	100% after 1000 h, at 35°C with 40% RH	NM	146
	TCO/NiO/PVK/PCBM/ZnO NPs/S-ALD ZnO/ITO	14.7	96% after 1500 h, at 65°C in N ₂	Negligible	152
	ITO/NiO _x /PVK/c-Nb ₂ O ₅ (001)/PC ₆₁ BM/Bphen/Ag	11.74	80% after 400 h, in N ₂ -filled glovebox at 85°C		137
	ITO/PEDOT:PSS/PVK/PCBM/AZO/SnO _x /Ag/SnO _x	11.5	① 100% after 4500 h, in N ₂ at 60°C	2.61	147
	ITO/PEDOT:PSS/PVK/PCBM/AZO/SnO _x /Ag	12.7	① ~90% after 1000 h, at 25°C with 60% RH ② ~100% after 3000 h, in N ₂ -filled glovebox at 60°C	Negligible	153

TABLE 1 (Continued)

Architecture	PCE (%)	Stability	Hysteresis (%)	Reference
ITO/PEDOT:PSS/PVK/PCBM/AZO/SnO _x /Ag	12.8	① 100% after 350 h, at 23°C with 50% RH ② ~100% after 1032 h, in N ₂ -filled glovebox at 60°C	0.55	148
FTO/NiO/PVK/PCBM/BCP/AZO/Ag	18.45	86.7% after 500 h, continuous 1 sun illumination at 85°C with 20%–60% RH	1.76	60
FTO/NiO _x /PVK/BaSnO ₃ /PCBM/Ag	16.2	92% after 30 days, nonencapsulated at ~20°C with ~30% RH	NM	46
ITO/NiO _x /PVK/PCBM/Zn ₂ SnO ₄ /Ag	17.76	~90% after 14 days, with 30% RH	NM	143
FTO/Cu:NiO _x /PVK/PC ₆₁ BM/CeO _x /Ag	17.35	90% after 1000 min, continuous illumination	Negligible	29
FTO/NiMgLiO/PVK/PCBM/CeO _x /Ag	18.7	① 100% after 200 h, in N ₂ -filled glovebox ② 91% after 200 h, under continuous light soaking with ~30% RH	0.64	34
ITO/PTAA/MAPI ₃ /PC ₆₁ BM/ZnO NPs/Al	17.2	NM	NM	215
FTO/NiO/PVK/PCBM/ZnO/Al	17.75	NM	NM	2
ITO/PEDOT:PSS/PVK/ZnO-PCBM/Ag	6.08	67% after 20 days, encapsulated, RT with 30% RH	2.14	3
ITO/NiO _x /PVK/PCBM/AZO/Ag	13.6	NM	NM	5
ITO/NiO _x /PVK/PCBM/SnO ₂ /Ag	18.77	NM	Negligible	207
ITO/PEDOT:PSS/PVK/PC[70]BM/AZO/Al	13.3	Over 90% after 170 h, exposure	Negligible	42
ITO/NiO/PVK/PC ₆₀ BM/AZO/TPPO/Ag	19.2	80% after 800 h, constant illumination	NM	6
ITO/NiO _x /PVK/PCBM/Nb-TiO ₂ /BCP/Ag	20.7	100% after 1000 h, 1 sun continuous illumination at open-circuit conditions in N ₂ at 35°C	0.49	71
ITO/PEDOT:PSS/PVK/PC ₇₀ BM/TiO ₂ /Al	14.03	100% after 23 days, with encapsulation in N ₂ glove box	NM	81
FTO/NiO _x /PVK/C ₆₀ /SnO ₂ -HClO ₄ /Ag	16.36	>80% after 90 days, in air with 55% ± 10% RH	4.6	48
ITO/NiO _x /PVK/PCBM/Ti(Nb)O _x /Ag	18.49	90% after 500 h, at 85°C with 85% RH	Negligible	155
ITO/NiO:BN/PVK/PCBM/ZnO/Ag	20.74	~85% after 60 days, in air with 30%–60% RH	NM	164
ITO/NiO _x /PVK/Eu ₂ O ₃ /PC ₆₁ BM/Bhen/Ag	14.09	>90% after 400 h, thermal aging at 80°C	NM	167
ITO/NiO _x /PVK/PCBM/CeO _x /Ag	17.47	85% after 300 h, at 30°C with 50% RH	4.92	168

(Continues)

TABLE 1 (Continued)

	Architecture	PCE (%)	Stability	Hysteresis (%)	Reference
	ITO/NiO/PVK/PCBM/ZnO/Ag	17.23	① >87% after 4500 h, unencapsulated, in N ₂ ② >82% after 200 h, encapsulated, at 85°C with 85% RH	0.5	165
	FTO/NiO _x /PVK/C ₆₀ /SnO ₂ -HClO ₄ /Ag	16.36	>80% after 90 days, in air with 55% ± 10% RH	4.6	48
	ITO/PEDOT:PSS/GO:PEG/PVK/PCBM/MoS ₂ /Ag	19.14	NM	NM	225
	ITO/CuI/PVK/PCBM/AZO/Ag	~14.21	~90% after 15 days, at ~35°C with ~50% RH	NM	170
	ITO/PTAA/WS ₂ /PVK/C ₆₀ /ZnSe/Cu(Ag)	20.92	78% after 40 days, at 25°C with ~30% RH	1.43	171
	ITO/PEDOT:PSS/PVK/PVK//PC ₆₁ BM/ZnMgO/Al	15.61	~90% after 20 day, in glove box	0.45	129
	ITO/PEDOT:PSS/WSe ₂ /PVK/PCBM/ZnO/Ag	16.2	29% after 30 days, Unencapsulated at 25°C with 30%–40% RH	NM	216
	ITO/PEDOT:PSS/PVK/CdSe/ZnS/C ₆₀ /BCP/Ag	20.00	75% after 2400 h	NM	172
	ITO/NiO _x /PVK/ZnO NPs/C ₆₀ /Ag	15.60	85% after 350 h, continuous light soaking in N ₂	NM	173
	FTO/Mo/Sb ₂ Se ₃ /PVK/C ₆₀ /GZO/Ag	16.8	NM	NM	222
	FTO/MoSe ₂ /PVK/PMMA/C ₆₀ /AZO/Ag	18.0	NM	NM	223
	ITO/NiO _x NP/PVK/PCBM/ZnO NP/Al	18.6	①87% after 1000 h, continuous light soaking(with UV filter) at 35°C in N ₂ ② > 80% after >1000 h, at 100°C	NM	213
	ITO/PVK/C ₆₀ /SnO ₂ /ITO	14.2	① 95% after 1000 h, unencapsulated in air in the dark at 85°C ②97% after 1000 h, encapsulated at 85°C with 85% RH	NM	1
	ITO/NiO _x /PVK/PC ₆₁ BM/SnO _x /Al	19.7	NM	2.03	219
Organic–inorganic hybrid ETL	FTO/NiO _x /PVK/ZnO@C ₆₀ /Ag	13.3	80% after 360 h, unencapsulated at 85°C	NM	61
	FTO/NiO _x /PVK/C ₆₀ :Co-TiO ₂ /Ag	22.13	① 50% after 110 h, continuous one-sun illumination in ambient air ② 97% after 70 days, 20%–87% RH	2.5	159
	ITO/NiO _x /PVK/TiO ₂ /PCBM:C ₆₀ /Ag	20.5	① ~90% after 350 h, continuous light soaking at 30°C with 50 ± 5% RH ② ~98.5% after 16 months, at 30°C with 50% ± 5% RH	1.95	64

TABLE 1 (Continued)

	Architecture	PCE (%)	Stability	Hysteresis (%)	Reference
	FTO/NiO/PVK/ALD Al ₂ O ₃ /PCBM/PPDIN6/Ag	20.40	95.3% after 150 days, unencapsulated in dry N ₂ glove box	Negligible	175
	ITO/NiO/PVK/PCBM-SnS ₂ /ZnO/Ag	19.95	80% after 50 days, in ambient atmosphere	14.27	226
	ITO/PEDOT:PSS/PVK/PC ₆₁ BM/ZnO@PFBC/Al	16.93	80% after 200 h, heating at 85°C in N ₂	NM	65
	FTO/NiO _x /PVK/ZnO@C ₆₀ /Ag	12.60	>90% after 10 h, unencapsulated under the stabilized power output operation	NM	174
	ITO/PEDOT:PSS/PVK/PCBM/BCP:ZnO NPs/Ag	15.50	NM	NM	217
	ITO/NiO/PVK/PC ₆₀ BM/Phen-NaDPO:Sn(SCN) ₂ /Ag	18.2	100% after 80 h, in dark (O ₂ and H ₂ ≤ 10 ppm)	0	227
	ITO/PEDOT:PSS/-PVK/Phen-NaDPO:Sn(SCN) ₂ /Al	18.2	NM	NM	227
	ITO/CuInS ₂ /PVK/PCBM/ZnO:TIPD/Ag	14.1	NM	3.55	126
All-inorganic ETLs	FTO/NiO/GO/PVK/GO-Li/TiO _x /Al	11.2	70% after 12 days, in a dry box in the dark 28%–32% RH	Negligible	31
	ITO/PEDOT:PSS/PVK/CdSe QDs/LiF/Ag	14.2	NM	NM	177
	ITO/c-NiO _x /mp-NiO _x /PVK/Zn(In)O/Al	16.2	>85% after 460 h, continuous light soaking at 30°C with 20–30% RH, encapsulated		145
	FTO/Cu:NiO _x /PVK/CdS/Au	13.36	NM	NM	228
	ITO/MoS ₂ /PVK/TiO ₂ /Ag	20.43	NM	NM	210
	FTO/NiMgLiO/PVK/c-Nb:TiO ₂ /Sb	14.8	① > 95% after 1000 h, dark at 85°C in N ₂ ② > 90% after 1000 h, under 1 sun in N ₂ at 60°C	3.2	158
	FTO/NiMgLiO/PVK/c-TiO ₂ /Bi/Ag	13.85	95% after 1000 h, encapsulated under 1 sun in N ₂ at 45°C	NM	62
	ITO/NiO/PVK/Bi ₂ S ₃ /Au	13.1	~80% after 30 days, unencapsulated at 25°C with 50%–75% RH	6.11	50
	ITO/NiO _x /PVK/ZnO/Al	16.1	~90% after 60 days, 25°C with 30%–50% RH	NM	72
	FTO/NiO _x /PVK/SnO _x /Al	13.5 ± 1.8	NM	NM	4
	FTO/GO/PVK/ZnO/Ag	1.35	NM	NM	161
	ITO/NiO _x /PVK/CeO _x /Ag	17.1	>90% after 200 h, in N ₂ at 25°C	4.09	154
	FTO/CZTS/PVK/ZnO/Al	3.48	80% after 72 h, in N ₂	9.09	162
	FTO/KFeO ₂ /PVK/ZnO/Pt	10.83	85% after 10 days, ambient environment at 30°C with 40%–45% RH	NM	163

(Continues)

TABLE 1 (Continued)

Architecture	PCE (%)	Stability	Hysteresis (%)	Reference
FTO/NiO _x /PVK/TiO _x /Al	13.3	>60% after 1050 h, uncover condition at 25°C with 45% RH	NM	166
Cu/CuI/PVK/ZnO/Ag	12.8	NM	NM	211
FTO/Mo/Cu ₂ ZnSnSe ₄ /PVK/ZnS/IZO/Ag	17.4	NM	NM	220
FTO/NiO _x /PVK/MoO _x /Au	5.52	long-term stability at high temperatures up to 160 °C	NM	45
FTO/Cu ₂ O/PVK/SiO ₂ /GZO/Ag	18.4	NM	NM	221
FTO/Cu:NiO _x /PVK/ZnO/Ag	16.51	90% after 6 months, unencapsulated under ambient conditions	0.18	169
ITO/NiO _x /PVK/CeO _x /Ag	5.60	90% after 500 h, Unencapsulated in an atmospheric condition at 40–45°C with 45%– 50% RH	NM	63
ITO/NiO _x /PVK/Sn:In ₂ O ₃ /In ₂ O ₃ /Ag	20.65	①91.9% after 69 day, under 85°C ②91.8% after 2000 h, 12 h continuous 1 sun illumination and then 12 h interval in the dark	0.7	203

Abbreviations: AZO, Al doped ZnO; BN, boron nitride; CZTS, Cu₂ZnSnS₄; GO, graphene oxide; GZO, Ga doped ZnO; IZO, In doped ZnO; NM, not mentioned; NP, nanoparticle; PVK, perovskite; RH, relative humidity; RT, room temperature.

are mainly used as inorganic top ETL. Among them, -ZnO,^{2,3,20,61,65,72,126,149,150,152,157,160–165,169,173,211–217} TiO_x,^{31,62,64,81,144,159,166,194,210,218} and SnO_x^{1,48,148,151,153,207,219} are the most popular choices. In addition, Al, In and Ga doped ZnO are also promising for electron transporting in inverted PSCs.^{5,6,42,60,121,147,148,153,170,220–223} The reported inorganic materials used as ETM are summarized in Table 1.

2.3.1 | Binary metal oxides and their modification

ZnO is a n-type semiconductor metal oxide with advantages of wide bandgap (3.3 eV), high exciton binding energy, low cost, low temperature fabrication, and high electron mobility. These excellent properties facilitate its application as efficient ETL in high performance inverted PSCs and benefit to the device commercialization. You et al. gained greatly enhanced air-stability of inverted PSC by using ZnO as top ETL. This ZnO based device nearly had no PCE decrease after 60 days in ambient air with room light illumination.⁷² For large-area durable flexible PSCs, ZnO was used as top ETL to get high PCE and stability.²¹¹ Recently, ZnO nanoparticles (NPs) were synthesized

through a low temperature (64.7°C) process.¹⁶¹ ZnO based device gained 10.83% PCE and retained 60% of its initial PCE after 250 h at 30°C with 40%–45% relative humidity.¹⁶³ Combined with the use of inorganic Cu:NiO_x HTL, ZnO based inverted PSC showed better PCE and excellent stability than the device based on conventional organic transport layers (Figure 3).¹⁶⁹

To further improve the properties of ZnO thus the performance of inverted PSC based on ZnO, different modification methods have been used. For metal oxides, doping is a conventional way to enhance their properties, especially the electronic characteristics. Based on semiconductor physics, doping element with higher valence than Zn into ZnO is beneficial for improving its electronic properties.²⁰⁴ This strategy has been adopted and doped ZnO has boosted the performance of PSCs based on this metal oxide. Al doped ZnO (ZAO) with better properties than pristine ZnO has been widely used as top ETL in inverted PSCs.^{5,6,42,60,121,147,148,153,170} Shown in Figure 4, Al doping enhanced the properties of pristine ZnO, such as transmittance, conductivity, and thus improve the device performance. Similarly, In References 151,220 and Ga^{221,222} have also been applied as dopants in ZnO and used as efficient top ETL in inverted PSCs.

TiO_x, nearly the original inorganic ETL used in n-i-p PSCs, has also been utilized for efficient electron transport in inverted PSCs.^{31,62,64,81,144,166,194,218} More suitable energy level and higher conductivity of C-TiO₂ than

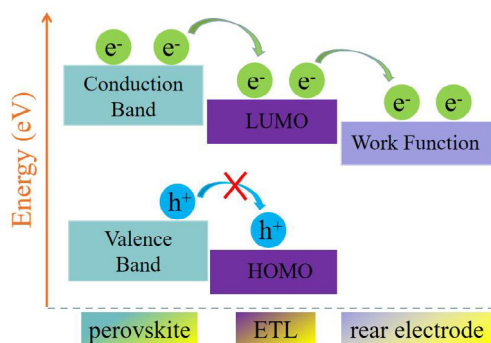


FIGURE 3 Diagram for charge transfer between perovskite layer, ETL, and rear electrode in inverted PSCs

C-In₂O₃, C-In₂O₃, C-SnO₂, and C-ZnO facilitated is as an excellent top ETL than the other four oxides (shown in Figure 5A,B). Ultrasmall 4 nm TiO₂ nanocrystals were synthesized and used as top ETL in inverted PSCs by Choy et al. and ~20% PCE, high shelf-life stability, and reproducibility were gained. TiO₂ also shows high transmittance like ZnO, and excellent electronic properties. According to the UV-vis absorption in Figure 5E, after coating TiO₂ on perovskite layer, nearly no light loss were observed. The device stability was greatly enhanced by comparing with the organic ETL based device.

As the same as ZnO, the properties of TiO_x can be adjusted by doping.²⁰⁴ As shown in Figure 6A, Nb doped TiO₂ boosted higher PCE and reduced hysteresis.¹⁵⁸ Additional electron in Nb⁵⁺ compared with Ti⁴⁺ enabled higher electron mobility.⁷¹ The enhanced properties made the ETL more conductive and the inverted PSC with higher PCE due to higher J_{sc} (Figure 6). Doping of transition metal Co and Fe tuned the electronic

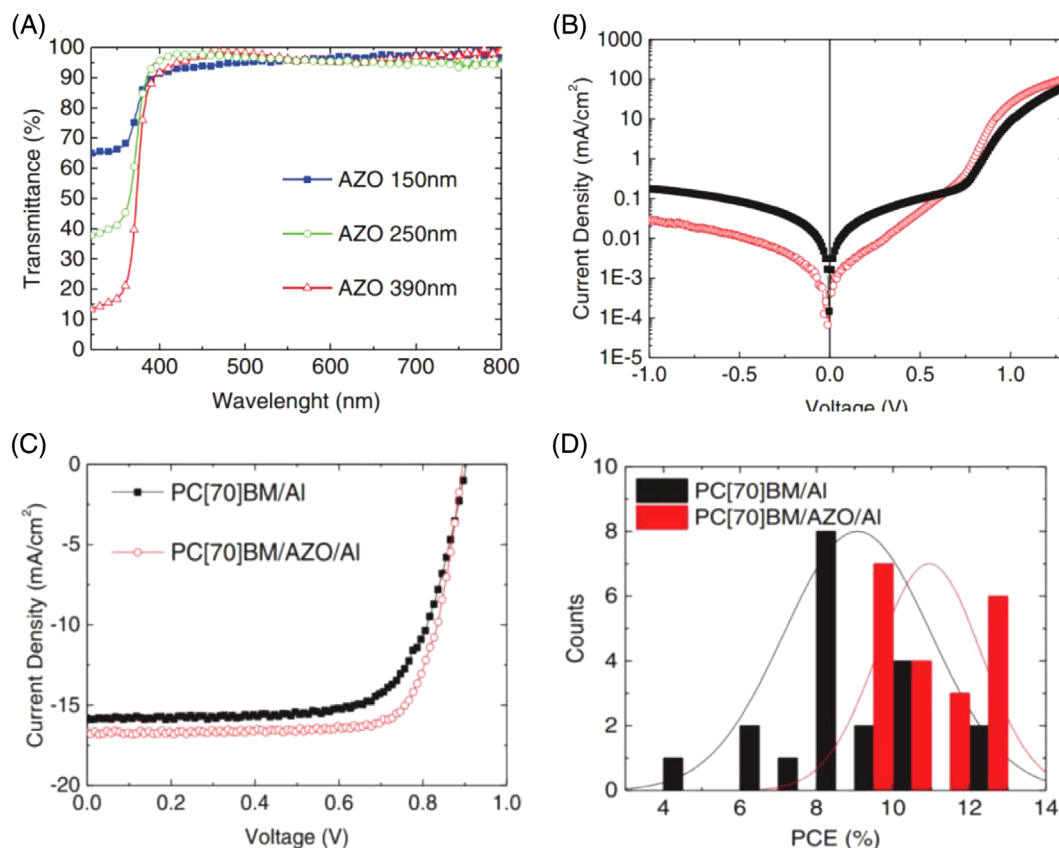


FIGURE 4 (A) Transmittance spectra of AZO layers with different thickness fabricated on quartz substrates, 150 nm (blue filled squares), 250 (green open circles), and 390 nm (red open triangles). (B) Current density versus voltage characteristics of two compared devices ITO/PEDOT:PSS/CH₃NH₃Pb(I_{1-x}Cl_x)₃/PC[70]BM/Al (black filled squares) and ITO/PEDOT:PSS/CH₃NH₃Pb(I_{1-x}Cl_x)₃/PC[70]BM/AZO/Al (red open circles) under dark conditions. (C) Current density versus voltage characteristics of two compared devices ITO/PEDOT:PSS/CH₃NH₃Pb(I_{1-x}Cl_x)₃/PC[70]BM/Al (black filled squares) and ITO/PEDOT:PSS/CH₃NH₃Pb(I_{1-x}Cl_x)₃/PC[70]BM/AZO/Al (red open circles) under illumination. (D) Distribution of the PCE represented in histograms as obtained from 20 pixels in two separated experimental runs.

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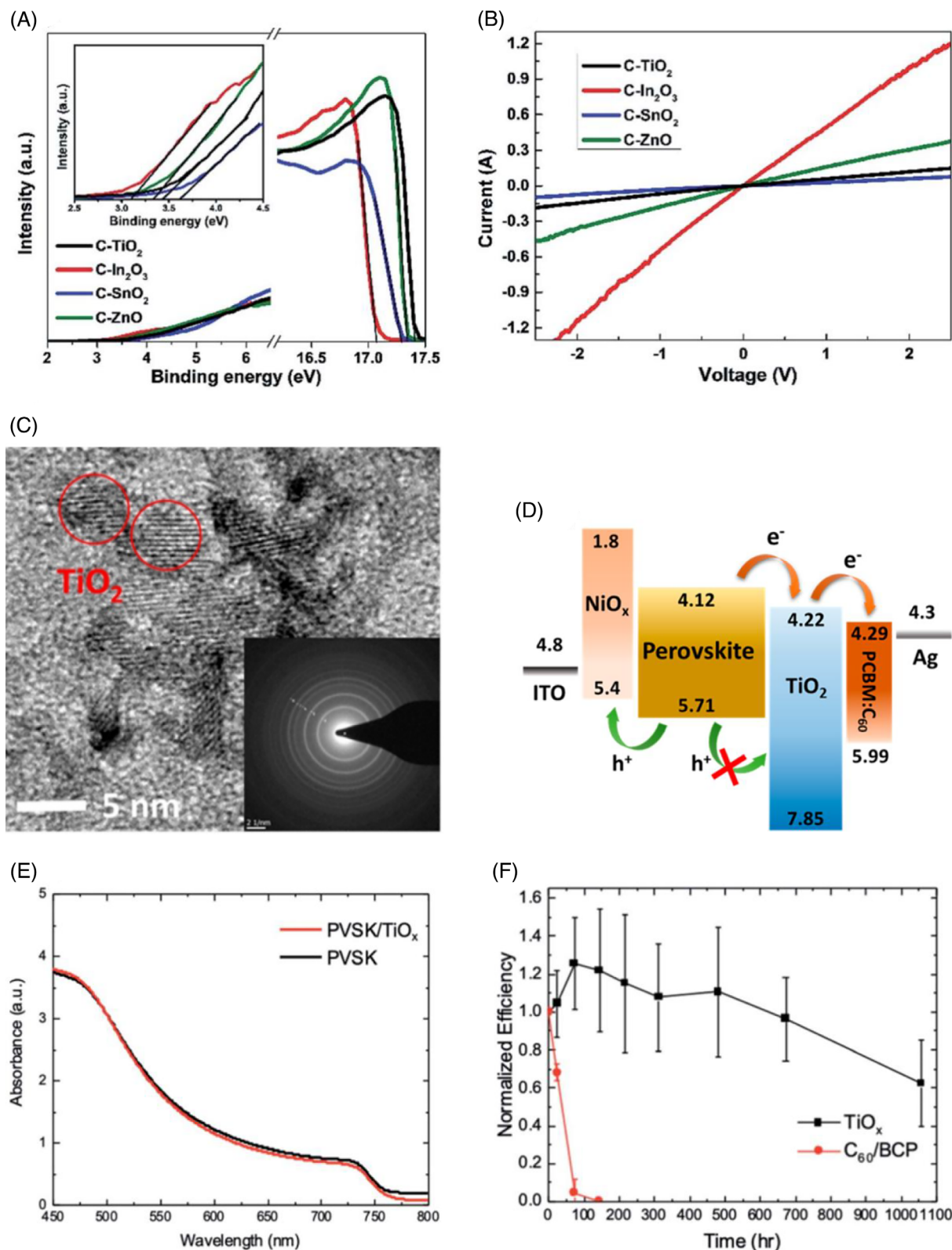


FIGURE 5 (A) UPS and (B) conductivity measurements for different carbide-metal oxide ETLs. (C) TEM image and the corresponding diffraction pattern of TiO₂ nanocrystals. (D) Corresponding energy band diagram of TiO₂-fullerene-based PSCs. (E) UV-vis absorption spectra before and after TiO_x deposition. (F) Normalized efficiency of devices with TiO_x ETL and C₆₀/BCP ETL shelf-aged in uncovered and ambient conditions over 1000 h, taken from eight devices over four substrates for each ETL.

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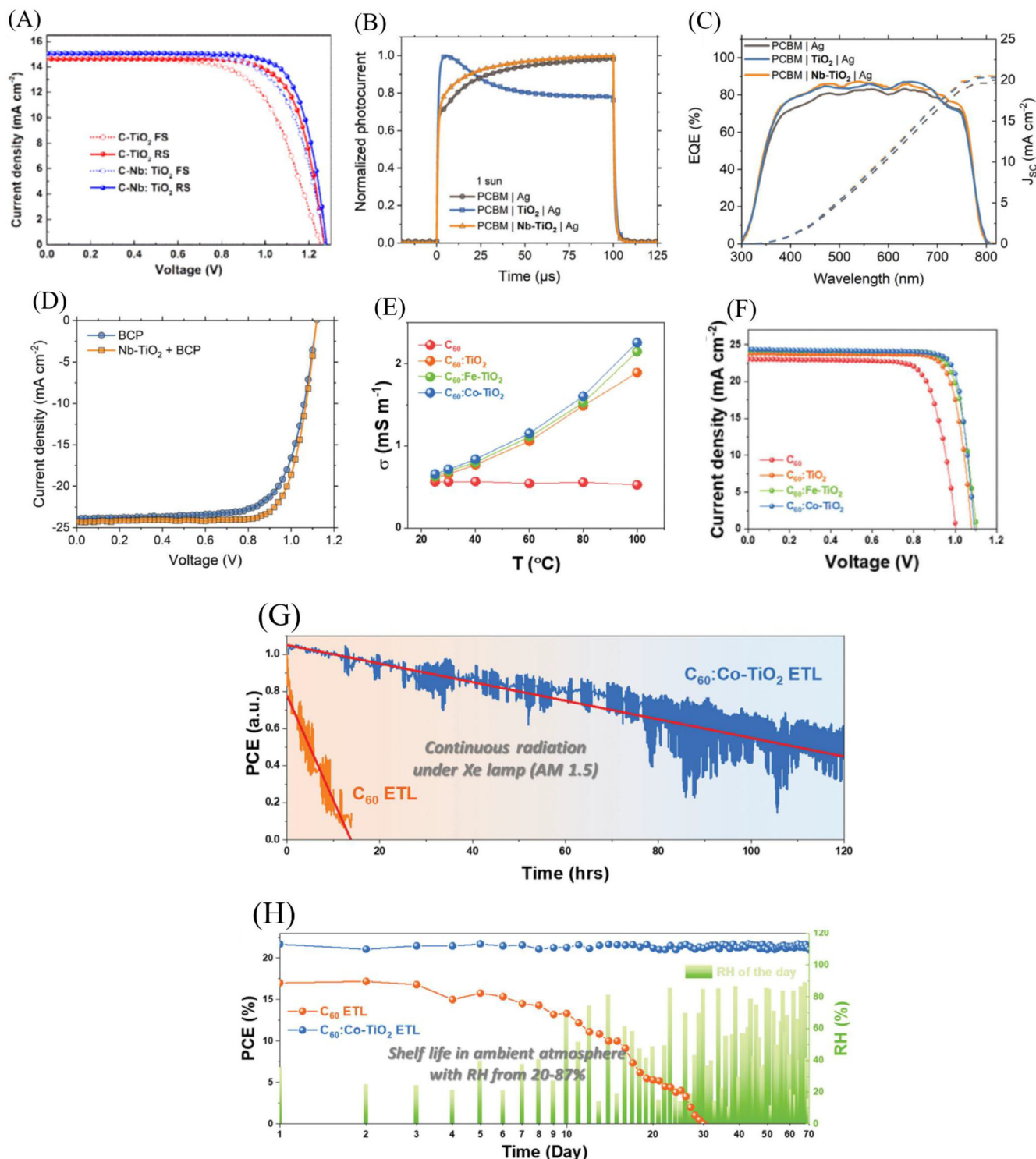


FIGURE 6 (A) J-V curves of PSCs with C-TiO₂ and Nb-doped C-TiO₂ ETLs. (B) Transient photocurrent measurements. (C) External quantum efficiency. (D) J-V curves from champion perovskite devices employing a PTAA HTL both with and without the interfacial bilayer. (E) The electrical conductivity was measured from devices of FTO/ETL/Al at different temperatures. (F) J-V curves of inverted PSCs using different ETLs. (G) Stability test under continuous one-sun Xe lamp illumination in ambient air, measured by tracking the current at MMP of an unencapsulated device for 120 h. (H) Device shelf life measurement by testing the J-V curve every day for over two months. Realtime relative humidity is also included.

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properties of TiO_2 brookite nanorod (NR).¹⁵⁹ Co and Fe doped TiO_2 NR improved film uniformity, electron extraction and transferability, energy level matching with perovskite, and moisture resistance. Finally, the inverted PSCs based on Co or Fe TiO_2 NR was boosted to higher PCE and stability, which are shown in Figure 6.

Inverted PSC based on a SnO_x layer with an optimized thickness of 60 nm as efficient top ETL achieved a 19.7% PCE value.²¹⁹ This pinhole-free and conformal SnO_x layer directly grown on perovskite was minimal structural damage to the underlying perovskite layer and enhanced the performance of PSC (Figure 7). The PSC performance has been found to be better than the organic ETL based devices.^{1,4,48,147,148,207,219} This means that SnO_x is also an efficient top ETL.

Besides the common binary metal oxides of ZnO , TiO_x , and SnO , other metal oxides, such as CrO_3 ,¹⁵⁶ CeO_x ,^{29,34,63,154,168} Nb_2O_5 ,¹³⁷ Eu_2O_3 ,¹⁶⁷ MoO_x ,⁴⁵ In_2O_3 ,²⁰³ SiO_2 ,²²¹ and Al_2O_3 ,¹⁷⁵ VO_x ²²⁹ have been used as top ETL in efficient inverted PSCs. From the SCLC plots in Figure 8 A and B, we can see that CeO_x based device has lower trap density than PCBM based device, suggesting better properties of inorganic metal oxide than organic PCBM. Furthermore, the stability of CeO_x based PSCs shown much higher stability than PCBM based devices under different conditions.

Besides doping strategy discussed above, some other ways to modify the binary metal oxide top ETL have been adopted. One efficient approach is using organic material to form hybrid or bilayer ETL. This can improve the electron transport ability and thus the performance of inverted PSCs.

2.3.2 | Ternary metal oxide

Because of extraordinary optical and electrical properties, some ternary metal oxides,²⁰⁵ like Zn(In)O ,¹⁴⁵ Ti(Nb)O_x ,¹⁵⁵ BaSnO_3 ,⁴⁶ Zn_2SnO_4 ,¹⁴³ and ZnMgO ,¹²⁹ have been used as efficient top ETL in inverted PSCs. Compared with pure PCBM, the ternary metal oxides block the erosion of humidity and ion migration from the perovskite to Ag

electrode more efficiently. This reveals the PSCs with high stability in both ambient and humidity conditions.

Compared with binary metal oxides, ternary metal oxides may have better stability due to their chemical structure, boosting the PSCs to higher stability. Additionally, the synthesis of ternary metal oxides is not complicated than binary metal oxides. Both the better stability and facile synthesis suggest that ternary metal oxides are excellent alternatives as efficient top ETL for inverted PSCs (Figure 9).

2.3.3 | Other inorganic top ETL

Besides metal oxides, metal chalcogenides have also been applied as top ETL in inverted PSCs and boosted high device performance. Bi_2S_3 ,⁵⁰ ZnS ,^{172,220} CdSe ,^{172,177} ZnSe ,^{171,224} CdS ,²²⁸ MoS_2 ,²²⁸ SnS ,²²⁶ Sn(SCN)_2 ,²²⁷ have been applied in high performance inverted PSCs. As the same as ZnO , other II-VI semiconductor metal sulfides and selenides have good optical and electrical characters. The fabrication of metal chalcogenide top ETL are similar to metal oxide ones without more post-treatments. In addition, they also have good chemical stability. These original properties make them as suitable alternatives to metal oxides as top ETL in inverted PSCs.

The main parameters of electronic properties of reported inorganic ETLs are summarized in Table 2. It can be seen that these inorganic ETLs have good properties, concluding suitable energy level matching with perovskite, high electron mobility, and conductivity. The above advantages are beneficial for efficient electron extraction and transporting.

3 | FABRICATION AND MODIFICATION OF INORGANIC TOP ETL

To obtain inorganic top ETL with excellent properties through facile fabrication is essential to PSC performance

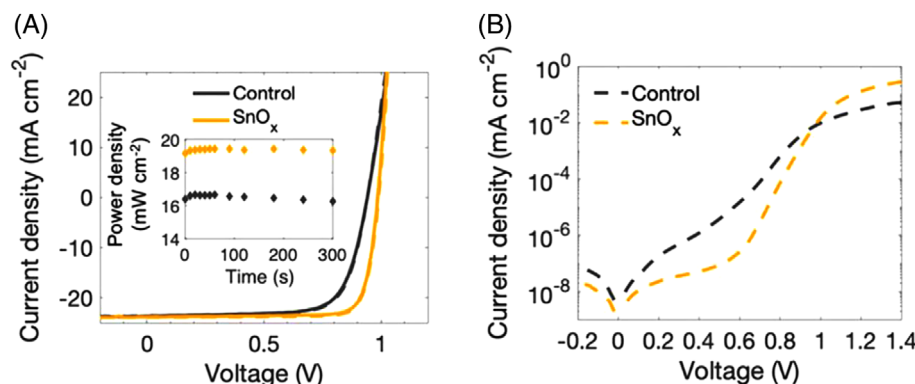


FIGURE 7 (A) Illuminated and (B) dark current-density versus Voltage curves and (inset) steady-state power output of triple-cation PSCs.

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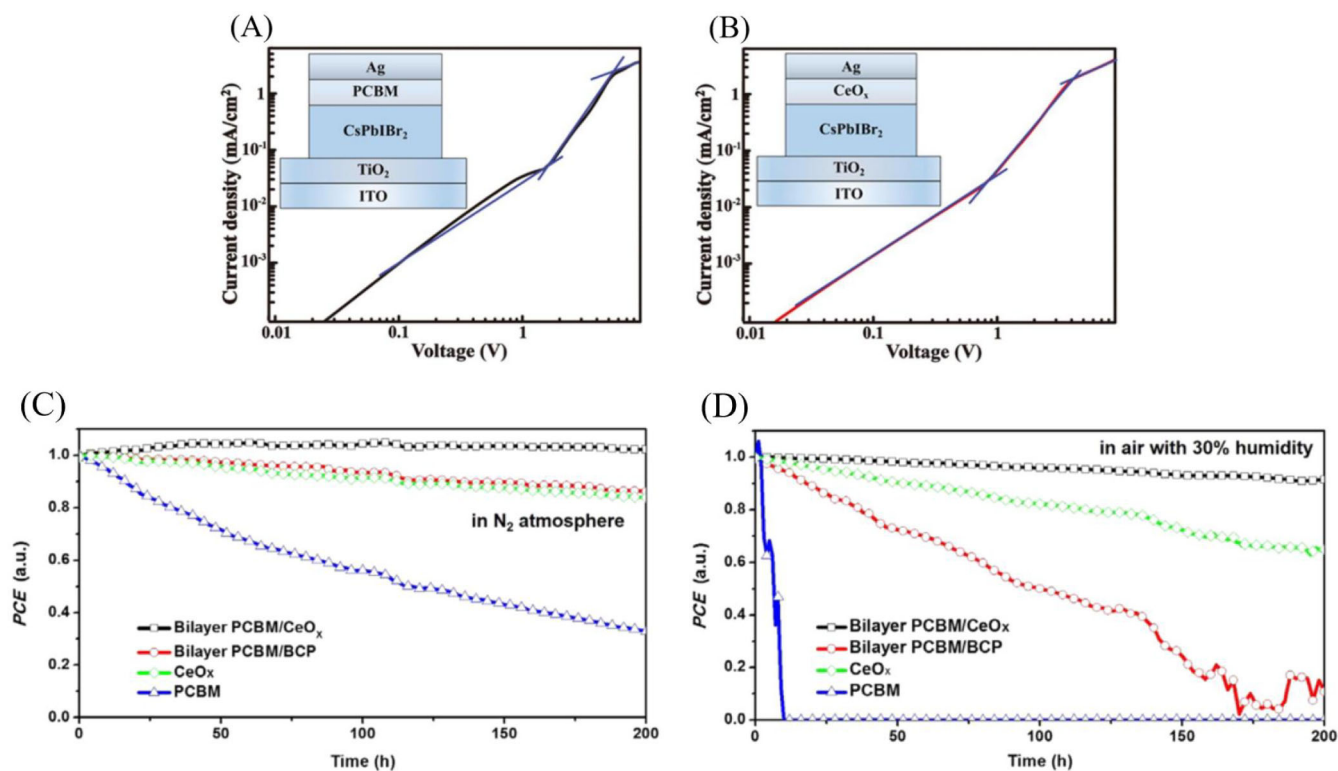
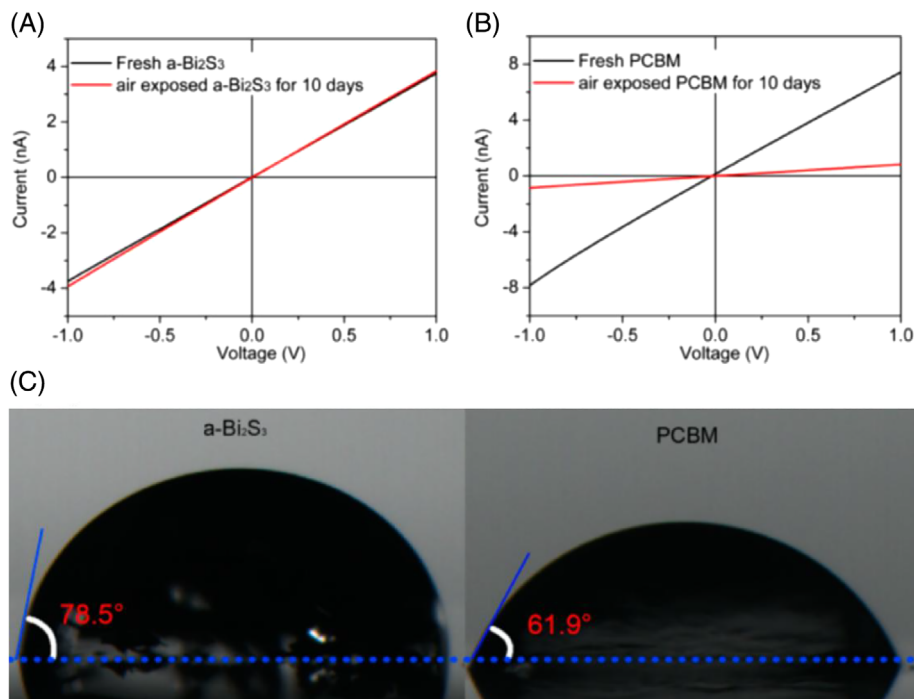


FIGURE 8 The space-charge-limited current (SCLC) plot of (A) ITO/TiO₂/perovskite/PCBM/Ag devices, and (B) ITO/TiO₂/perovskite/CeO_x/Ag devices. (C), (D) Normalized PCE of PSCs based on different ETLs under different conditions.

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FIGURE 9 Stability of Bi₂S₃ and PCBM films exposed to ambient air for 60 days.

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and its commercialization. In some cases, only pristine metal oxide does not show the optimal state as top ETL. So various modifications have been done to pristine

metal oxides to improve their properties for efficient inverted PSCs. We review the fabrication and modification of inorganic top ETL in this section.

TABLE 2 Electronic properties of some inorganic top ETL

	CB (eV)	VB (eV)	Mobility (cm ² /V·s)	Conductivity (S/cm)	Reference
ZnO	4.1	7.5	$\sim 10^{-5}$ – 10^2	$\sim 10^{-6}$	16,161
TiOx	3.9	7.65	$\sim 10^{-4}$ –1	$\sim 10^{-8}$ – 10^{-6}	16
SnOx			$\sim 10^{-8}$ – 10^2	$\sim 10^{-6}$	16
CeOx		7.69			154
Nb ₂ O ₅	3.94	7.0	4.97×10^{-4}		137
Eu ₂ O ₃	3.88	7.3			167
MoOx	4.0	7.8			45
Al ₂ O ₃			7.9		16
In ₂ O ₃	4.51	8.32	14–226		203
Ti(Nb)Ox	4.0	7.6			16
BaSnO ₃	3.91	7.11	150		230
Zn ₂ SnO ₄	4.1	7.9	10–30		230
ZnMgO	3.18	6.88			129
ZnS	–3.5	–7.1			220
Bi ₂ S ₃	3.86	5.45	257		50

3.1 | Fabrication

The top ETL fabrication process is an important step during the whole procedure of inverted PSC preparation. Different from n-i-p PSCs, ETL of inverted PSCs should experience relatively low temperature (commonly <110 °C) because high temperature will damage the perovskite film under ETL. Therefore, low-temperature or no post-treatment will prevent the perovskite from thermal degradation. Additionally, facile fabrication strategy can save cost and boost the commercialization of PSCs. The main fabricating methods to gain top ETL with high quality are as follows.

3.1.1 | Spin-coating of pre-synthesized nanoparticles

The most used method of top ETL forming is presynthesis and a following spin-coating. Inorganic ETMs for inverted PSCs are mostly synthesized through solvothermal method. In Figure 10 A, a typical synthesis of ZnO NPs is shown. This solvothermal method may bring in some insulated ligands such as oleic acid. Ligand exchange is needed before the spin-coating on perovskite film. After this exchange shown in Figure 10 B,C, ETM solution are dropped on the perovskite layer and then form as ETL by spin-coating shown in Figure 10D. To avoid the ligand exchange, solvent without long carbon chain can be used for NP synthesis. The adoption of a new solvent such as water or ethanol simplifies the fabrication process of NPs before spin-coating. Furthermore, the reaction temperature using oleic acid is higher than that of water and ethanol, suggesting low-cost procedure. The thickness of ETL formed by spin-coating

can be controlled through the concentration of ETL solution, spin time, spin speed and circles. Equally important, the synthesis and ligand exchange also affect the properties of top ETL and thus the performance of the inverted PSCs.

3.1.2 | Atomic layer deposition

To passivate perovskite surfaces, a promising way of atomic layer deposition (ALD) had been applied to form metal oxide.¹⁷⁵ Another strong point of ALD is that it is a scalable vapor-phase thin-film deposition strategy to gain highly uniform layer on rough perovskite surface with precisely controllable thickness and prominent reproducibility. The properties of metal oxide from by ALD can be changed with different temperature. Considering the drawback of classical ALD, spatial ALD was introduced to deposit efficient metal oxide in efficient inverted PSC.^{152,153} The schematic of the spatial ALD for top ETL is shown in Figure 11 A. This process can be finished under low temperature (80 °C), protecting the perovskite film below.

3.1.3 | Direct annealing strategy

Similar with the popular method to fabrication of ETL in n-i-p PSCs, top ETL for inverted PSCs can also be formed by annealing the precursor solutions. The process is shown in Figure 11B, suggesting a typical procedure of this strategy. Some things must be paid attention as follows: (a) low temperature below 110 °C to prevent the perovskite from thermal damage, (b) appropriate metal precursor to void insulated residue during low-temperature annealing

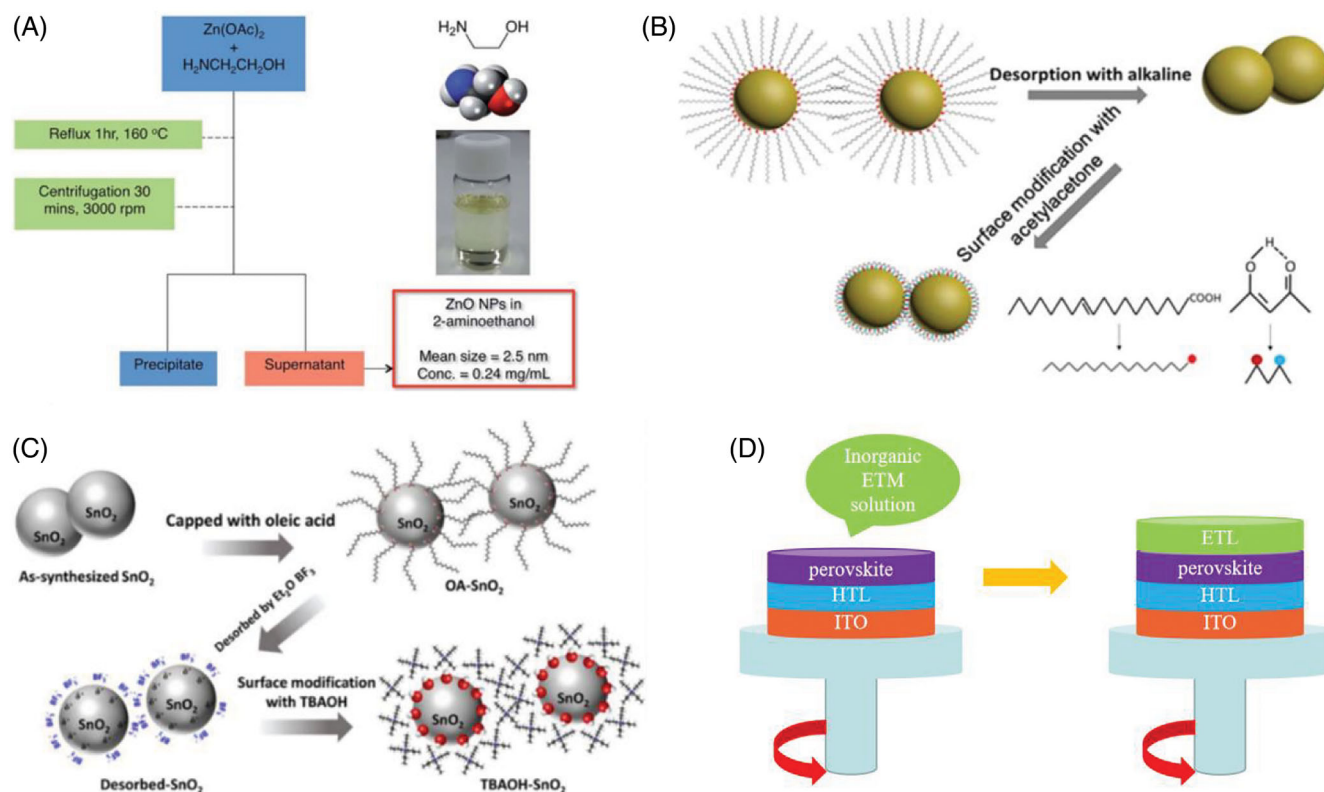


FIGURE 10 (A) Synthesis and separation of ZnO nanoparticles. Schematic illustrating the ligand exchange procedures (B) to convert oleic acid (OA)-capped CeOx NCs into actylacetone-modified ones, (C) for TBAOH-capped SnO_2 nanoparticles. (D) Schematic illustration of top ETL fabrication from pre-synthesized inorganic ETM.

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process, (c) optimized precursor concentration to gain trimmed thickness for pin-hole free and conductive ETL.

ETL can be precisely controlled and the optimized value will gain for efficient PSC.

3.1.4 | Chemical vapor deposition

Another alternative for growing metal oxide layer on perovskite film is atmospheric pressure chemical vapor deposition (AP-CVD). The illustration is shown as Figure 11C,D. We can see that the oxide is grown by a soft chemical-based way, which is unlike sputter-deposition. Furthermore, the growth rates are very high and no vacuum is required, which is unlike ALD and thermal evaporation and saves fabrication cost.

3.1.5 | Sputtering

Metal oxides can also be deposited on perovskite layer by magnetron sputtering. High energy ions bombardment during sputter can bring in low roughness of metal oxide layer, which induces good electric contact with the perovskite film. Similar with CVD method, the thickness of top

3.1.6 | Thermal evaporation

For inorganic materials as Bi_2S_3 , it can be deposited by simple thermal evaporation, which is controlled by tuning the current of a tungsten filament. This method has the same advantage as sputtering, well controlling the thickness of top ETL. Some more inorganic top ETL can be gained by this efficient method and used in high-performance inverted PSCs.

4 | SUMMARY AND OUTLOOK

Considering the shortcomings of organic ETLs, we surveyed and analyzed inorganic top ETL for efficient inverted PSCs in this review. From the reported inorganic top ETLs, we sum up their advantages compared with organic ETLs, which concludes facile fabrication, high electron conductivity, tunable energy level, high stability,

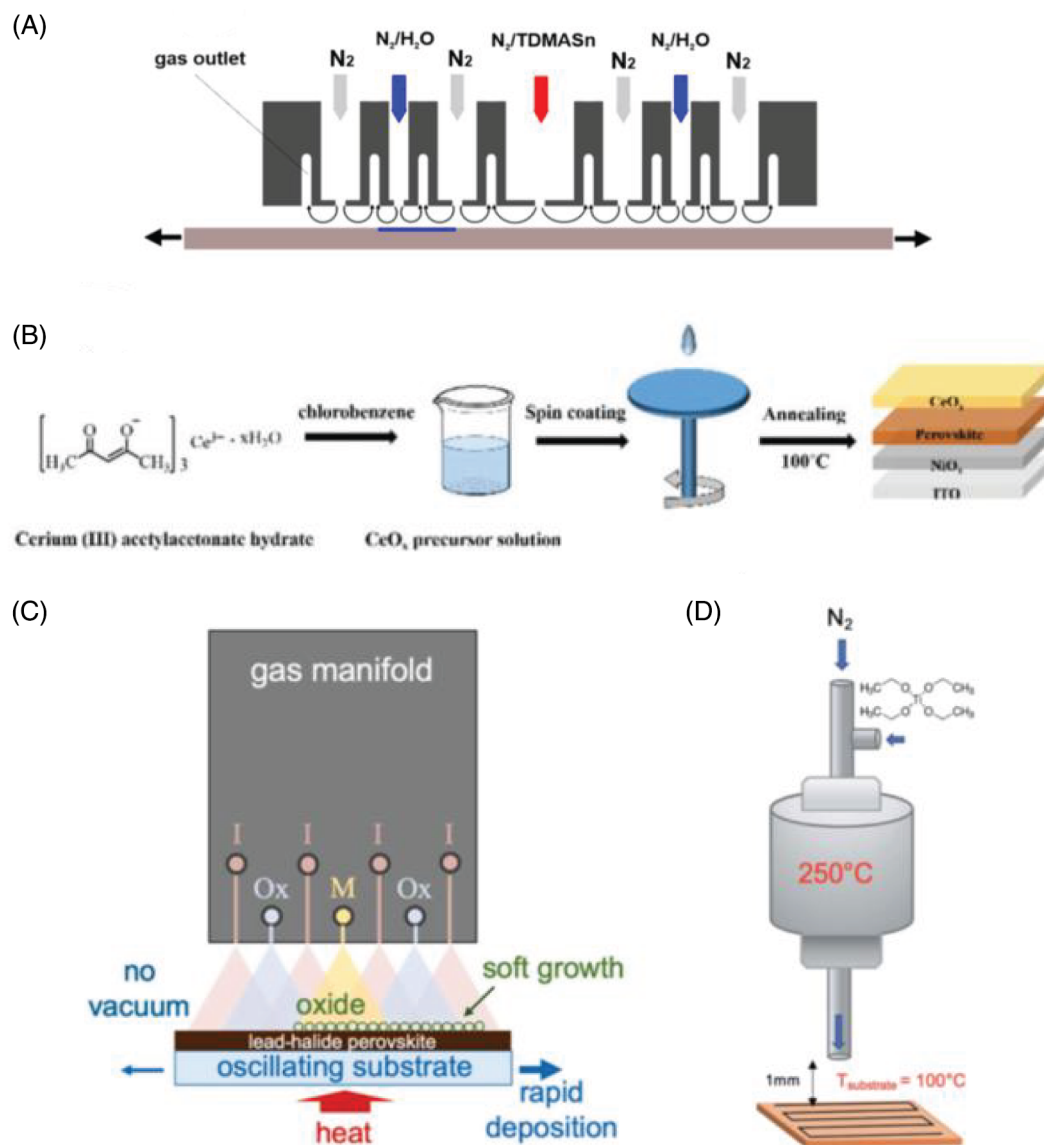


FIGURE 11 (A) Schematic of the spatial ALD assembly for SnOx. (B) Schematic diagram of the CeOx ETL fabrication. (C) Illustration of growing oxides on lead-halide perovskite film by AP-CVD. (D) OA-CVD setup with titanium ethoxide, flash evaporator, and deposition raster pattern.

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adjustable properties, and low cost. Although many inorganic materials have been used for efficient electron transporting in inverted PSCs, there is still much room to be deeply probed for further enhancement of ETL properties thus the performance of inverted PSCs. In the near future, efforts are anticipated to be thrown into, but not limited to, the following directions:

A. Finding more new inorganic ETLs for efficient inverted PSCs. Previous inorganic ETLs mainly focus on binary metal oxides, narrowing the range of

materials choosing. Multiple metal oxides, metal sulphide, and selenides with homologous properties can expand the inorganic ETL family and boost the PSC performance.

B. Exploring facile fabrication strategy. To simplify the procedure of device preparation, facile fabrication strategy of top ETL is beneficial to scalable deposition of top ETLs, which is promising for the commercialization of PSCs.

C. Improving the properties of inorganic ETLs. As known, ETL is a very important functional layer in

inverted PSCs. Improved properties of ETL is a direct and useful way to enhance device performance.

In summary, these efforts above will boost the PSC performance and the real application of the devices.

ACKNOWLEDGMENTS

The authors acknowledge the Natural Science Foundation of the Jiangsu Higher Education Institutions of China (20KJB430023), Qing Lan Project of Jiangsu Province (2021-11), the University Grant Council of the University of Hong Kong for the funding (Grant 201811159147 and 2019157209), the General Research Fund from the research Grants Council of Hong Kong Special Administrative Region (Grant 17204117, 17200518, 17201819 and 17211220), the Collaboration Research Fund (CRF Project C7035-20G) from the Research Grants Council (RGC) of Hong Kong Special Administrative Region, China, and Guangdong-Hong Kong-Macao Joint Laboratory for Photonic-Thermal-Electrical Energy Materials and Devices (2019B121205001).

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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How to cite this article: Yang B, Peng S, Choy WCH. Inorganic top electron transport layer for high performance inverted perovskite solar cells. *EcoMat.* 2021;3(5):e12127. <https://doi.org/10.1002/eom2.12127>