

Supporting Information

Optimized NiO_x deposition for Industrial Compatible Perovskite Solar Modules on 15 × 15 cm²: a Green Route Towards Stable Methyammonium-free Devices

Farshad Jafarzadeh^a, Luigi Angelo Castriotta^{a,*}, Emanuele Calabrò^{a,b}, Pierpaolo Spinelli^c, Francesco Di Giacomo^{a,d}, Fabio Matteocci^a, Francesca Brunetti^a, Aldo Di Carlo^{a,e}

Table S1. The comparison of Perovskite Solar Cell and Modules Fabricated using NiO_x as HTL in terms of active-area and PV characteristics.

Device Architecture	NiO _x Deposition	Deposition Environment	Active Area (cm ²)	PCE (%)	Ref.
FTO/NiO _x /CsFAMA Perovskite/C ₆₀ /BCP/Cu	Spray coating	N ₂	0.09	23.61	1
FTO/NiO _x /FAPbI ₃ /PCBM/PCB/Ag	Evaporation	N ₂	0.09	23.4	2
ITO/NiO _x /CsMAFAPb(IBr) ₃ /C ₆₀ /BCP/Au	Evaporation	N ₂	0.1	21.6	3
FTO/NiO _x /perovskite/C ₆₀ /BCP/Cu	Spray Coating	Air	0.1	20.7	4
FTO/NiO _x /CsFAPbI ₃ /C ₆₀ /BCP/Cu	Spray Coating	Air	0.1	19.5	5
FTO/NiO _x /MAPI/C ₆₀ /BCP/Ag	Blade coating	Air	0.12	19.01	6
FTO/NiO _x /CsFAPbI ₃ /PCBM/BCP/Ag	Spin Coating	N ₂	0.1	18.9	7
ITO/NiO/CsFAPbBrI ₃ /C ₆₀ /BCP/Ag	Spray Coating	N ₂	0.2	17.7	8
FTO/NiO _x /CsFAPbI ₃ /PCBM/BCP/Cu	Slot-die	Air	0.14	17.33	9
FTO/NiO _x /CsFAPbI ₃ /PCBM/BCP/Ag	Spin Coating	N ₂	1	16.2	7
ITO/NiO _x /CsMAFAPb(IBr) ₃ /C ₆₀ /BCP/Au	Evaporation	N ₂	0.1	15.6	10
ITO/NiO/CsFAPbI ₃ /C ₆₀ /BCP/Ag	Flexography	N ₂	0.134	15.3	11
FTO/NiO _x /CsFAPbI ₃ /PCBM/BCP/Cu	Slot-die	Air	2.1	14.9	9
FTO/NiO _x /MAPbI ₃ /PC ₆₁ BM/TBAOH/Ag	Slot-die	Air	0.09	13.86	12
FTO/NiO _x /MAPbI ₃ /PC ₆₁ BM/TBAOH/Ag	Slot-die	Air	0.75	13.54	12
ITO/NiO _x /CsMAFAPb(IBr) ₃ /C ₆₀ /BCP/Au	Evaporation	N ₂	2.3	12.4	10
FTO/NiO _x /MAPI/C ₆₀ /BCP/Ag	Blade coating	Air	25	11.07	6
FTO/NiO _x /MAPI/PCBM/PEI/Ag	Slot-die	Air	0.09	10.96	13
FTO/NiO _x /MAPI/PCBM/PEI/Ag	Slot-die	Air	3.78	10.34	13
FTO/NiO _x /CsFAMA Perovskite/C ₆₀ /BCP/Au	Chemical Bath Deposition	N ₂	28.0	19.29	14
FTO/NiO _x /SAM/Perovskite/PTAA/Ag	Spin coating	N ₂	60.84	17.04	15
ITO/NiO _x /CsFA Perovskite/LiF/C ₆₀ /BCP/Cu	Evaporation	N ₂	100	15.5	16
FTO/NiO _x /FAPbI ₃ /PCBM/PCB/Ag	Evaporation	N ₂	156	18.6	2
FTO/NiO _x /CSFAMA Perovskite/C ₆₀ /BCP/Cu	Spray Coating	N ₂	178.4	17.18	1
FTO/NiO_x/SAM/CsFAPbBrI₃/C₆₀/BCP/Cu	Blade coating	Air	110	12.6	This Work

Table S2. The average thickness of doctor bladed NiOx based on the concentration of precursor ink based on the ellipsometry data.

Concentration	Avg. Thickness $\pm$ Std.
0.15 M (Ref)	39.40 $\pm$ 2.42
0.075 M (1:1)	42.18 $\pm$ 1.32
0.050 M (1:2)	40.05 $\pm$ 2.37
0.037 M (1:3)	36.20 $\pm$ 1.11

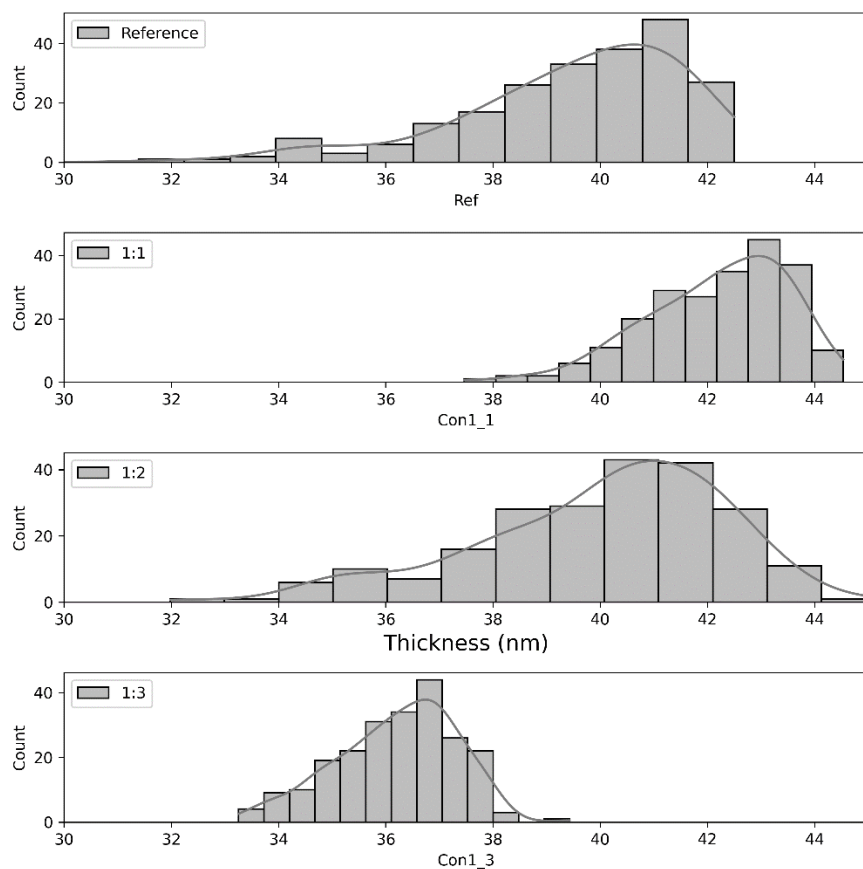


Figure S1. The thickness distribution of doctor bladed NiOx films by variation of precursor ink based on the ellipsometry data.

References

1. Li, C. *et al.* Efficient inverted perovskite solar cells with a fill factor over 86% via surface modification of the nickel oxide hole contact. *Adv. Funct. Mater.* **33**, 2214774 (2023).

2. Du, M. *et al.* Surface redox engineering of vacuum-deposited NiO_x for top-performance perovskite solar cells and modules. *Joule* **6**, 1931–1943 (2022).
3. Eggers, H. *et al.* Inkjet-printed micrometer-thick perovskite solar cells with large columnar grains. *Adv. Energy Mater.* **10**, 1903184 (2020).
4. Zhang, X. *et al.* Blade coating inverted perovskite solar cells with vacuum-assisted nucleation based on bottom quasi-2D passivation. *Sol. RRL* **7**, 2200900 (2023).
5. Zhang, X. *et al.* Blade-coated inverted perovskite solar cells in an ambient environment. *Sol. Energy Mater. Sol. Cells* **246**, 111894 (2022).
6. Solar, P. *et al.* Stabilized and Operational Pbl₂ Precursor Ink for Large-Scale Perovskite Solar Cells via Two-Step Blade-Coating. *J. Phys. Chem. C* **124**, 8129–8139 (2020).
7. Li, N. *et al.* Zwitterion-Stabilizing Scalable Bladed α -Phase Cs_{0.1}FA_{0.9}PbI₃Films for Efficient Inverted Planar Perovskite Solar Cells. *ACS Sustainable Chemistry and Engineering* **8**, 7020–7030 (2020).
8. Scheideler, W. J., Rolston, N., Zhao, O., Zhang, J. & Dauskardt, R. H. Rapid aqueous spray fabrication of robust NiO_x: A simple and scalable platform for efficient perovskite solar cells. *Adv. Energy Mater.* **9**, 1803600 (2019).
9. Le, T. S. *et al.* All-slot-die-coated inverted perovskite solar cells in ambient conditions with chlorine additives. *Sol. RRL* **6**, 2100807 (2022).
10. Abzieher, T. *et al.* Electron-beam-evaporated nickel oxide hole transport layers for perovskite-based photovoltaics. *Adv. Energy Mater.* **9**, 1802995 (2019).
11. Huddy, J. E., Ye, Y. & Scheideler, W. J. Eliminating the perovskite solar cell manufacturing bottleneck via high-speed flexography. *Adv. Mater. Technol.* **7**, 2101282 (2022).
12. Li, C.-F. *et al.* High-performance perovskite solar cells and modules fabricated by slot-die coating with nontoxic solvents. *Nanomaterials (Basel)* **13**, 1760 (2023).

13. Huang, S.-H. *et al.* Toward all slot-die fabricated high efficiency large area perovskite solar cell using rapid near infrared heating in ambient air. *Adv. Energy Mater.* **10**, 2001567 (2020).
14. Liao, Y. *et al.* Seed-assisted Cu-doped chemical bath deposition for preparing high-quality NiO_x hole-transport layers in perovskite solar cells. *Sol. RRL* **7**, (2023).
15. *Co-assembled Monolayers Modified Nickel Oxide for High Efficient Perovskite Solar Cells and Modules.*
16. Tutundzic, M. *et al.* Toward efficient and fully scalable sputtered NiO_x-based inverted perovskite solar modules via co-ordinated modification strategies. *Sol. RRL* (2023)
doi:10.1002/solr.202300862.