Supporting Information

Air-processed efficient and pure green perovskite LEDs based on PVP-modified NiO_X interface layer

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Experimental Methods

Materials: Cesium bromide (CsBr, 99.999%) and dimethyl sulfoxide (DMSO, 99.8%) were purchased from *Alfa Aesar, Thermo Fisher Scientific Inc*. Formamidinium bromide (FABr, 99.5%), phenethylammonium bromide (PEABr, 99.5%), 2,2',2''-(1,3,5-Benzinetriyl)-tris(1-phenyl-1-H-benzimidazole) (TPBi, 99.9%) and poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS, 4083) were purchased from *Xi'an Yuri Solar Co., Ltd*. Lead bromide ($PbBr_2$, 99.999%), 18-crown-6 (Crown, 99%) and triphenyl phosphorus oxide (TPPO, 98%) were purchased from *Sigma-Aldrich*. Ethyl acetate (EA, 99.5%) was purchased from *Shandong Keyuan Biochem*. Polyvinylpyrrolidone (PVP, M.W. 40000) was purchased from *Aladdin Scientific Corp*. Pattern ITO (indium tin oxide) glass (15×15 mm, active area was 2×2 mm) with a sheet resistance of ~15 Ω sq^{-1} were purchased from *Advanced Election Technology Co., Ltd*. Nickel(II) nitrate hexahydrate ($Ni(NO_3)_2•6H_2O$, 98%) and sodium hydroxide (NaOH, ≥96%) were purchased from *Sinopharm Chemical Reagent Co., Ltd*. All materials were used without any pre-treatment.

Preparation of perovskite precursor: The precursor solution was prepared by mixing CsBr, FABr, $PbBr_2$, PEABr and Crown into DMSO solvent with molar ratio of 0.04:0.2:0.08:0.18:0.014. The mixture was stirring at 50 °C in nitrogen filled glovebox until all powders dissolved, forming a transparent precursor solution. The perovskite precursor was stored in glovebox and filtrated (PTFE-0.22 μm) each time before use.

Fabrication of NiO_x nanocrystals: 29 g $Ni(NO_3)_2•6H_2O$ were dissolved into 20 mL deionized water to obtain dark green solution. Then the pH value of the solution was regulated to 10 by adding 400 mg mL^{-1} NaOH aqueous solution. After 15 min stirring, the mixture was vacuum filtrated. The green participates were washed by deionized water for three times and dried in vacuum at 80 °C for 6 h. The green powders were then calcined in muffle furnace at 270 °C for 2 h and black NiO_x nanocrystals were acquired. The as-prepared NiO_x nanocrystals were stored in N_2 glovebox.

PeLED fabrication: Patterned ITO glass was rinsed in detergent, deionized water and ethanol with sonication for 30 min respectively. NiO_x nanocrystals were dissolved in deionized water with concentration of 30 mg mL^{-1}, and the solution was filtrated (PES-0.22 μm) before use. NiO_x (40 μL), deionized water (100 μL) and PVP (4 mg mL^{-1} in DMF, filtrated by PES-0.22 μm, 25 μL) were subsequently spin-coated on the cleansed substrate. The spin-coating was carried out at 4000 rpm for 30 s, 5000 rpm for 30 s and 5000 rpm for 30 s respectively. The substrate was then annealed at 120 °C for 5 min. For the deposition of perovskite layer, the perovskite precursor solution on the as-prepared substrate at 4000 rpm for 1 min. TPPO in EA solution (5 mg mL^{-1}, 40 μL) was added as anti-solvent at 25 s and then the perovskite film was annealed at 70 °C for 5 min. All the procedures above were accomplished in the air with humidity of 50 ± 5%. Then the substrate was transfer in glovebox to evaporate 40 nm of TPBi, 0.8 nm of LiF and 100 nm of Al subsequently, using a high-vacuum evaporator. The final device structure was ITO/PEDOT:PSS/NiO_x/PVP/Perovskite/TPBi/LiF/Al. For the hole transport layer (HTL) only device, the TPBi/LiF/Al layers were substituted by MoO_3 (20 nm)/Au (50 nm) through high-vacuum evaporation.

Film and device characterization: Atomic force microscopy (AFM) images were collected by Park NX10. X-ray diffraction (XRD) was measured by Rigaku MiniFlex. UV absorption and transmittance were measured by Hitachi UH4150. Contact angle was measured by Aifeisi FCA2000A (China). Time-resolved photoluminescence (TRPL) was measured by Horiba Fluorolog-QM, with 405 nm pulse laser excitation. PL spectra, PL quantum yield (PLQY) and J-V-L-EQE were measured by XPQY-EQE-Adv (*Xi Pu Optoelectronics Technology Co., Ltd.*). Scanning electron microscopy (SEM), X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) were measured by a third testing agency, *Shanghai WEIPU Testing Technology Group Co., Ltd.*

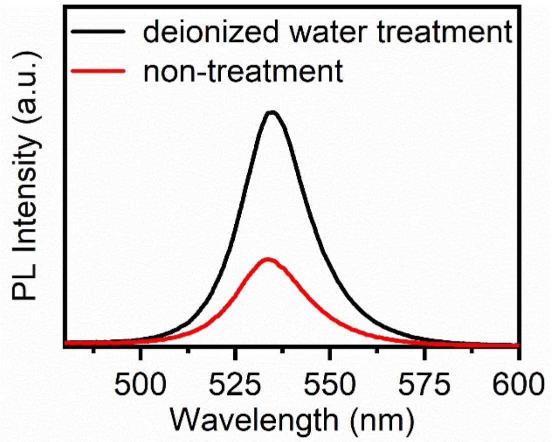

Figure S1. PL spectra of perovskite coating on the non-treated and deionized water-treated NiO_x.

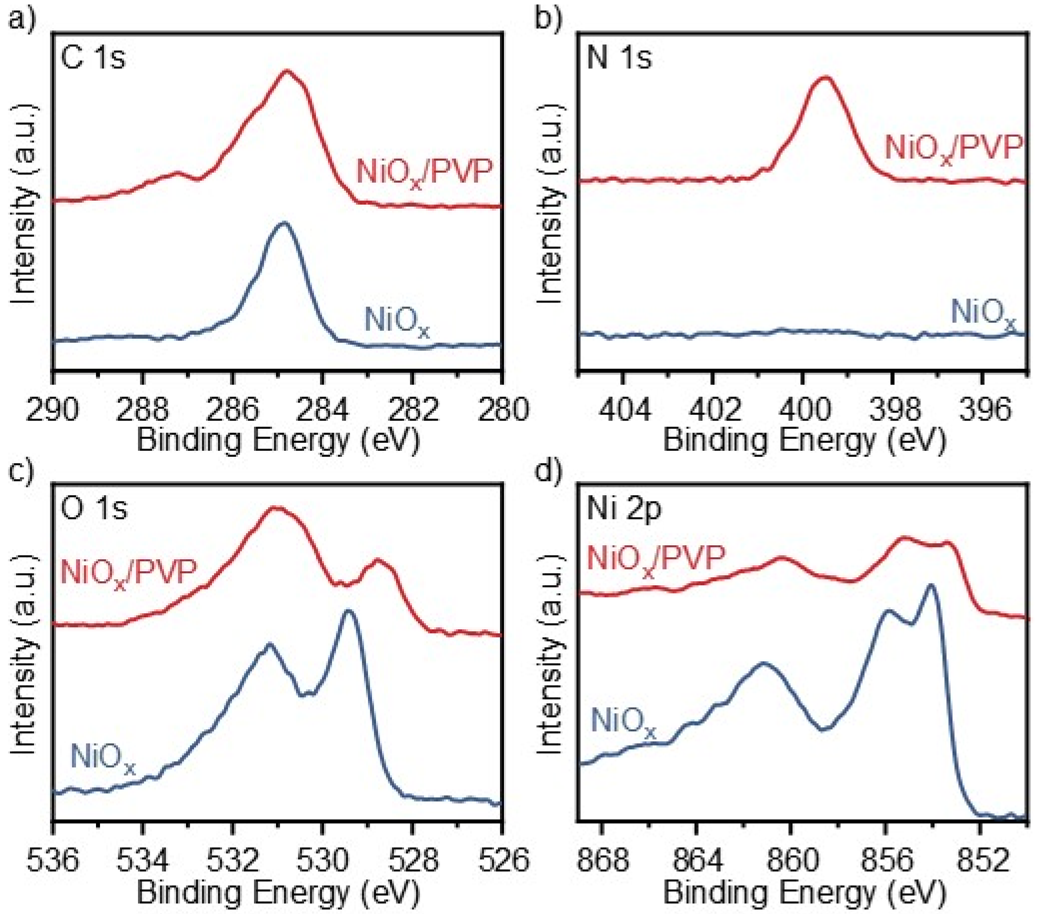

Figure S2. XPS spectra of NiO_x and PVP modified NiO_x. C_{1s} (a), N_{1s} (b), O_{1s} (c) and Ni_{2p} (d) spectra.

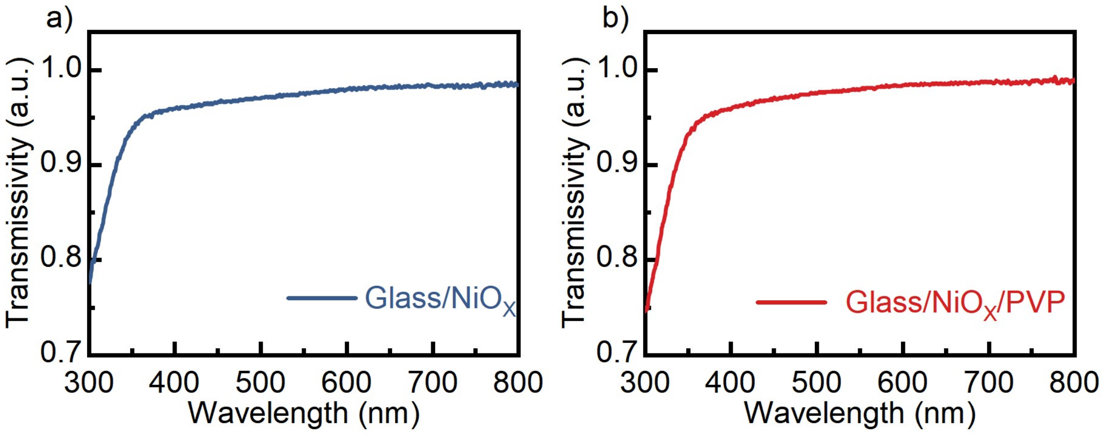

Figure S3. Transmittance of NiO$_x$ on glass (a) and PVP-modified NiO$_x$ on glass (b).

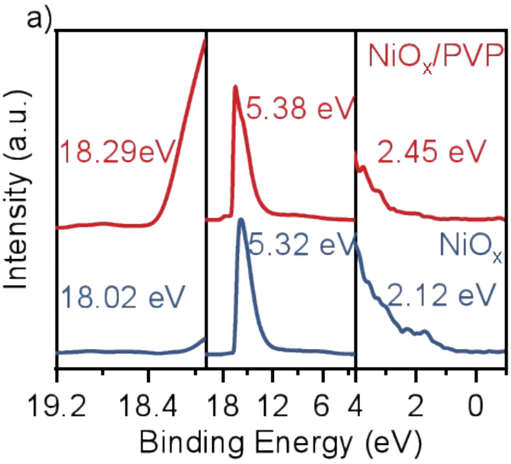

Figure S4. UPS spectra of NiO$_x$ and PVP modified NiO$_x$.

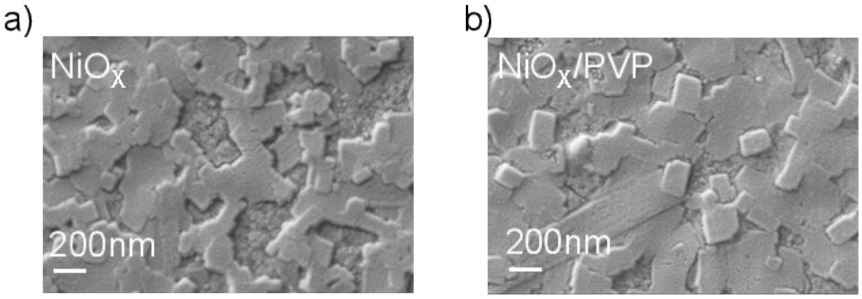

Figure S5. SEM images of perovskite layers deposited on NiO_x (a) and PVP-modified NiO_x (b).

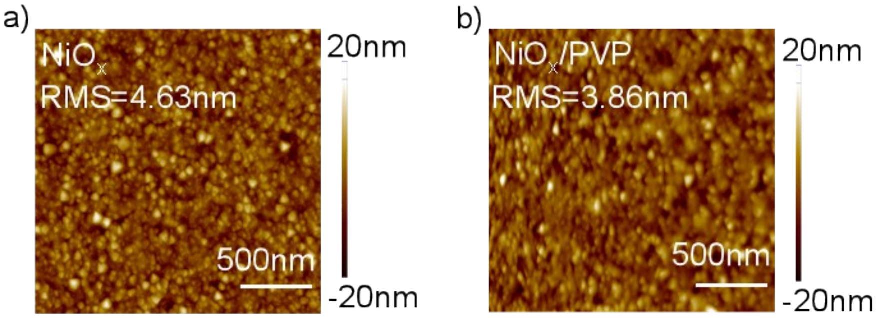

Figure S6. AFM images of perovskite layers deposited on NiO_x (a) and PVP-modified NiO_x (b).

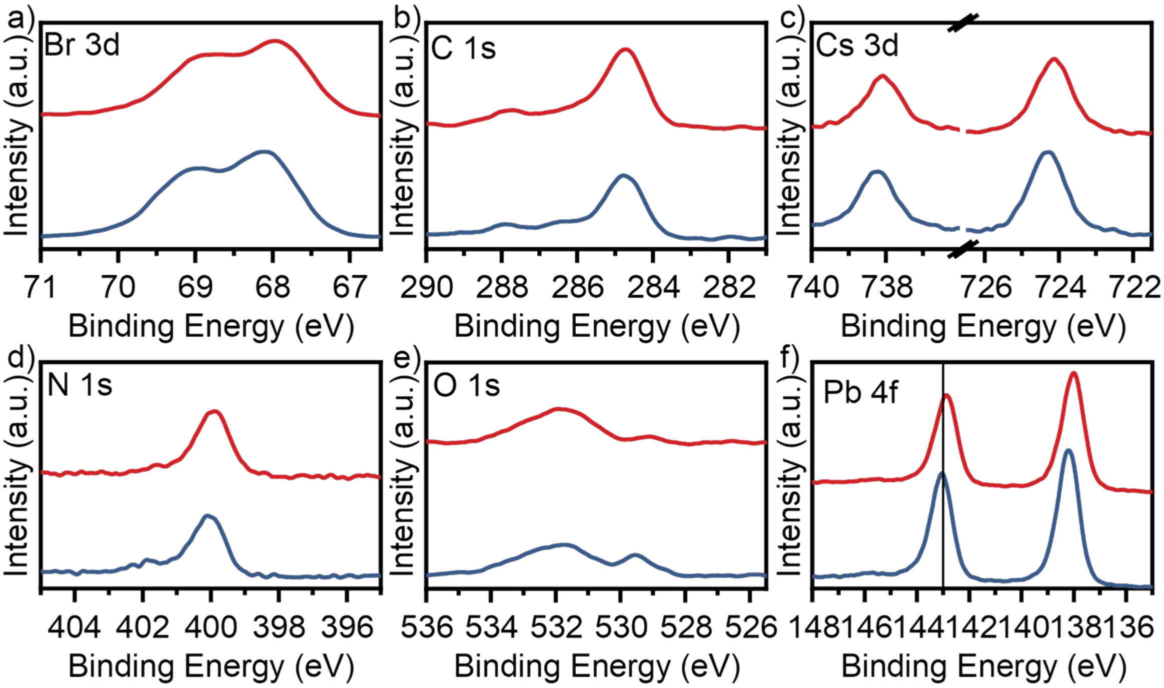

Figure S7. XPS spectra of perovskite thin films deposited on NiO_x and PVP-modified NiO_x. Br 3d (a), C 1s (b), Cs 3d (c), N 1s (d), O 1s (e)and Pb 4f (f) spectra.

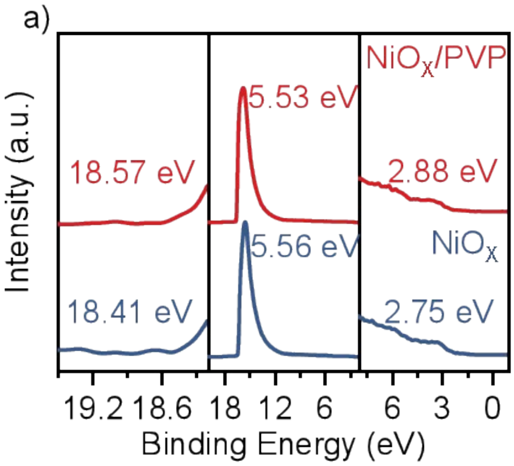

Figure S8. UPS spectra of perovskite layers deposited on NiO$_x$ and PVP-modified NiO$_x$.

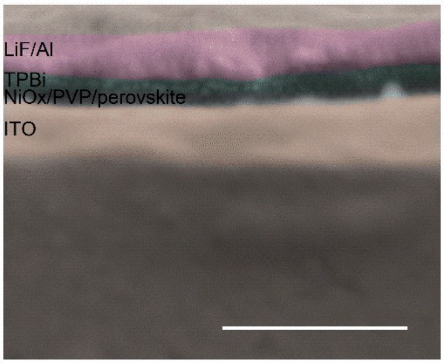

Figure S9. Cross-sectional image of PeLED device, the scale bar is 250 nm.

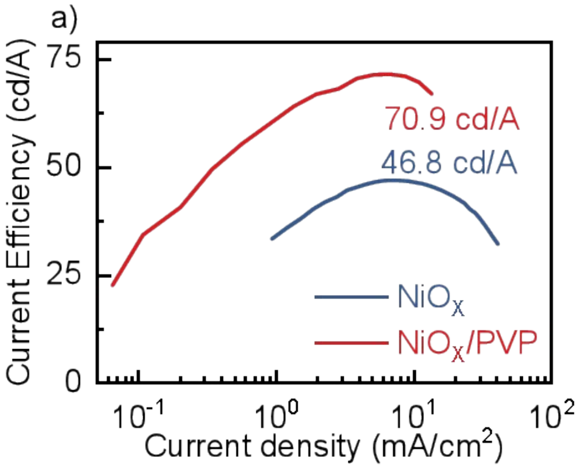

Figure S10. Current efficiency as a function of current density of two devices.

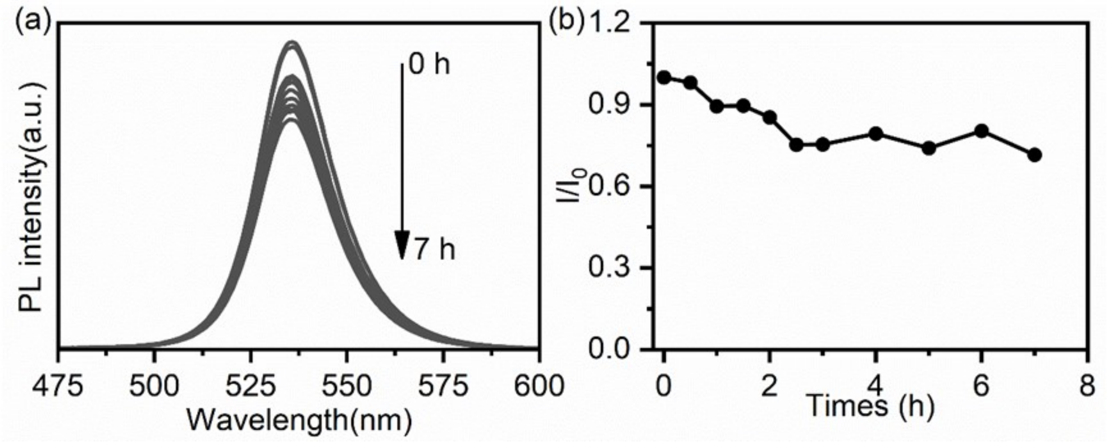

Figure S11. PL attenuation in air, (a) PL spectra obtained after different exposure time in air, (b) I/I_0 as a function of exposure time in air. The PL intensity maintains ~70% of its initial value after 7 hours of continuous air exposure.

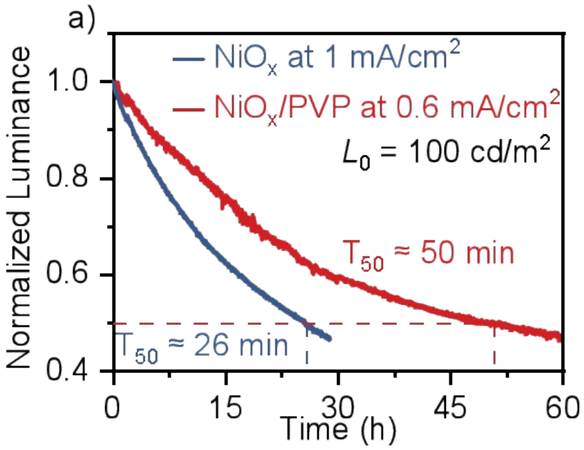

Figure S12. Lifetime measurements of the two devices operating at luminance of 100 cd/m^2.

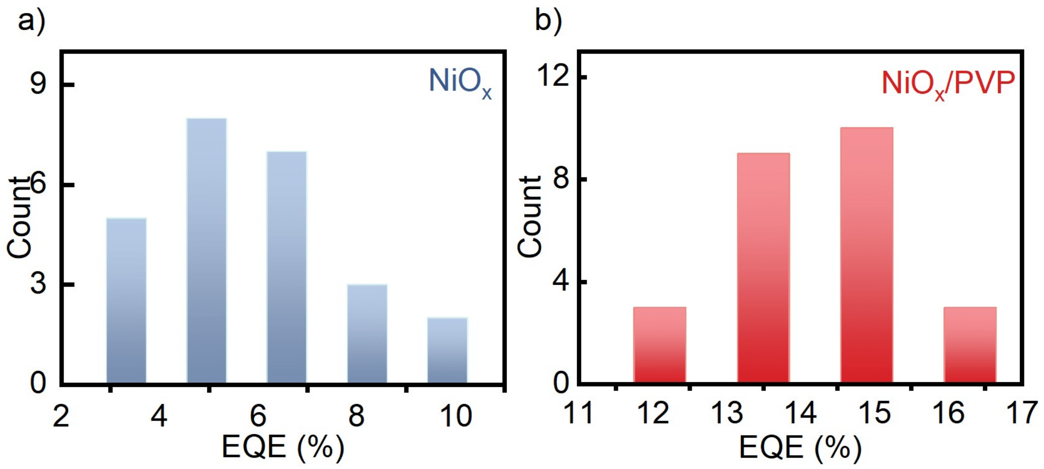

Figure S13. EQE statistics of 25 devices with NiO$_x$ and NiO$_x$/PVP as HTL respectively.

Table S1. Bi-exponential fitting results for TRPL spectra of the perovskite films prepared with various hole transport layer.

	A_1 (%)	τ_1 (ns)	A_2 (%)	τ_2 (ns)	$\tau_{average}$ (ns)
NiO_x	99.8	6.99	0.2	87.22	4.85
NiO_x/PVP	57.3	76.45	42.7	647.1	52.99

Table S2. Summary of device parameters of NiOx and PVP-modified NiOx based PeLEDs.

	V_T (V)	EQE_{max} (%)	L_{max} (cd/m^2)	CE_{max} (cd/A)	EL (nm)	FWHM (nm)	CIE (x, y)
NiO_x	2.9	10.55	15856	46.8	535	21	(0.22, 0.75)
NiO_x/PVP	2.8	16.05	12004	70.9	535	21	(0.22, 0.75)

Table S3. Summary of reported air processed green PeLEDs.

HTL	Device Architecture	Emission Wavelength (nm)	EQE (%)	Reference
PEDOT:PSS	ITO/PEDOT:PSS/PEA-$FAPbBr_3$/TPBi/LiF/Al	525	15.4	1
PEDOT:PSS	ITO/PEDOT:PSS/$MAPbBr_3$/PMMA/TPBi/Ag	539	0.16	2
2PACz/PFNBr	ITO/2PACz/PFNBr/$CsPbBr_3$/TPBi/LiF/Al	513	12.06	3
2PACz	ITO/2PACz/$CsPbBr_3$/TPBi/LiF/Al	521	4.87	4
PEDOT:PSS	ITO/PEDOT:PSS/PEA-$CsPbBr_3$/Ag	520	0.35	5
NiO_x/PVP	ITO/NiO_x/PVP/PEA-$FAPbBr_3$/TPBi/LiF/Al	535	16.05	This Work

References

1. Liu et al, *Adv. Funct. Mater.*, **2021**, 31, 2103399.
2. Kim et al, *Phys. Status Solidi A*, **2019**, 216, 1900642.
3. Li et al, *Adv. Funct. Mater.*, **2023**, 33, 2303787.
4. Li et al, *ACS Appl. Mater. Interfaces*, **2023**, 15, 36602-36610.
5. Cheng et al, *Phys. Status Solidi RRL*, **2023**, 17, 2200295.