Effect of surface modification of nickel oxide hole-transport layer via self-assembled monolayers in perovskite solar cells

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1 INTRODUCTION

Since the last decade, the field of perovskite solar cell (PSC) has been at the forefront of the research of photovoltaics. For the first time in 2009, the perovskite material was used in a solar cell device by Miyasaka et al., yielding an efficiency of 3.8%. Since then, there have been numerous attempts to improve the efficiency and the stability of PSCs, with the toxicity of the lead-based perovskite material as more recent concern. 

For enhancing device efficiency, various approaches have been considered, for instance, changing deposition condition for improved morphology or changing active layer composition for more light absorption or reduced defect sites. 

Besides active layer engineering, other approaches include introducing additional layers to improve the charge transfer towards the respective electrode. The additional layer could be another charge transport layer, or a surface modification layer to adjust the interfacial energy level alignment. Self-assembled monolayers (SAMs) have been widely used as interface modifiers in various electronic devices. Specifically, in PSCs, SAMs have also been used, though more often in the n-i-p device configuration, and considerable improvements in the performance were reported. The SAM formation process typically involves immersion of the substrate in a solution of modifying molecules. In the n-i-p configuration, the SAM is formed on the surface of electron-transporting layer, such as TiO₂, to realize its effect on the device performance.
There are reports that the SAM modification influences the morphology of the perovskite layer. For instance, Zuo et al. reported a pin-hole-free perovskite film on a C3-SAM (3-aminopropanoic acid)-modified ZnO substrate when compared with an unmodified ZnO underlayer.\cite{24} There are also reports where the SAM modification did not have any effect on the morphology of the perovskite film.\cite{15} Hence, depending on the functionality of the SAM used, the effect of SAM modification can be varied, influencing the morphology/crystallinity or the energy level alignment, or both.

In the cells with p-i-n configuration, the commonly used hole-transporting layer (HTL) poly(3, 4-ethylenedioxythiophene)poly(styrene sulfonate (PEDOT: PSS) is acidic in nature and relatively unstable. The PEDOT:PSS is suggested to degrade the device performance.\cite{25–27} Metal oxide layers as charge transport layers have shown promising improvement over their organic counterparts and specifically, in case of p-i-n configuration, nickel oxide-based HTL have been shown to have advantages in terms of stability and device performance.\cite{28–33} These metal oxide layers can be readily modified through functional monolayers.\cite{34,35} Among the possible modifiers, phosphonic acids are known to anchor sturdily on metal oxides as compared to benzoic acid, silanes etc.\cite{36–38} Wang et al. modified the NiO$_x$ layer with a series of benzoic acid SAMs and observed an improved device performance in 4-bromobenzoic acid-based device.\cite{35} However, they reported a large change in perovskite film morphology/crystallinity as the impact of the SAM modification. It is difficult to delineate whether the improved device performance is a result of better band alignment or changed perovskite morphology/crystallinity, or even different thickness as a result of varied surface wettability after the SAM modification. As the device power conversion efficiency is a composite result of open circuit voltage ($V_{oc}$), short circuit current ($J_{sc}$) and fill factor (FF), which can all be affected by the morphology/crystallinity of perovskite and its contact with other layers and in turn be modulated by SAM modification, minimizing the number of variables involved may lead to better correlation.\cite{39,40}

Here in this report, we modified the nickel oxide surface with a series of para-substituted phenylphosphonic acid SAMs as the HTL in the fabrication of PSC to investigate their effect on the device performance. The different substituents (electron-donating CH$_3$O-, H- and electron-withdrawing CN- groups) impart different molecular dipoles to the molecules so that when anchored to the NiO$_x$ surface, the work function (WF) of the NiO$_x$ HTL can be tuned with respect to the valence level of the perovskite layer. The substituents are chosen such that the hydrophilicity of the modified surfaces is not much different, so that their effect on other variables such as perovskite film thickness and crystallinity upon deposition can be minimized. Their effects on the device performance were evaluated and correlation with respect to the band alignment of NiO$_x$ or charge extraction was studied.

2 | EXPERIMENTAL SECTION

2.1 | Materials

All chemicals used in the study are analytical grade. The indium tin-oxide (ITO) substrates were purchased from UNI-ONWARD Corp. The precursors for nickel oxide, nickel acetate tetrahydrate (Ni (CH$_3$CO$_2$)$_2$•4 H$_2$O, 99+%) and ethanolamine (HOCH$_2$CH$_2$NH$_2$) were purchased from Alfa Aesar and Fisher Scientific, respectively. The perovskite precursors, lead iodide (PbI$_2$) (metal basis 99.999%, ultra-dry) was obtained from Alfa Aeser and methylammonium iodide (CH$_3$NH$_3$IO or MAI) was obtained from Greatcell Solar. Dimethylformamide (DMF), dimethyl sulfoxide (DMSO) and chlorobenzene (CB) were purchased from Sigma Aldrich. PCBM was purchased from Lumtec. Phenylphosphonic acid (PPA) and 4-cyanophenylphosphonic acid (CNPPA) were synthesized in the lab. The 4-methoxyphenylphosphonic acid (MPPA) was obtained from TCI.

2.2 | Solar cell fabrication

2.2.1 | NiO$_x$ preparation

The NiO$_x$ was deposited by a slightly modified procedure from that reported previously.\cite{32,41} Thus, the precursor solution for nickel oxide was prepared by mixing 1 M equivalent of ethanolamine and nickel acetate tetrahydrate in ethanol and stirred for 24 hours at 50°C, after which a homogeneous dark-green solution was formed. The solution was filtered using 0.45 µm PVDF filter. ITO substrates (1.5 cm × 1.5 cm) were cleaned step-by-step with detergent solution, DI H$_2$O, ethanol, acetone in sequence, followed by UV-ozone treatment for 25 minutes. The NiO$_x$ precursor solution was spin-coated on ITO substrates at 3000 rpm for 30 seconds. The substrates were annealed at 250°C, for 1 hour and then cooled to room temperature before further process.

2.2.2 | SAM preparation

The NiO$_x$-ITO substrates were immersed in 1 mM ethanol solution of different phosphonic acid molecules at 50°C for
60 minutes. The substrates were then dried with N₂ gun before further heated at 150°C for 3 hours to ensure covalent bond formation. Then the substrates were let to cool down naturally till room temperature, thoroughly rinsed with ethanol to remove any physisorbed multilayers and dried with N₂ gun.

2.2.3 | Device fabrication

The device fabrication was carried out in a N₂-filled glove box. Equimolar amounts of PbI₂ (461 mg), MAI (159 mg), and DMSO (72 µL) were dissolved in 630 µL of DMF and stirred for 8 hours at 70°C. The clear golden-yellow solution was filtered using 0.22 µm PVDF filter and spin-coated on the pristine or SAM-modified NiOₓ substrate at 4000 rpm for 25 seconds. Two hundred microliters of CB was dripped at 8-11 seconds on top of the substrate after the onset of perovskite precursor spinning. The substrate was then annealed at 70°C for 1 minute and at 100°C for next 5 minutes. A homogenous perovskite film has a shiny black texture. The next step was to spin-coat the electron transport layer. A 2% PCBM solution in CB (stirred at 70°C for 8 hours) was spin-coated at 3000 rpm for 30 seconds and the substrate was subjected to annealing at 100°C for 10 minutes. A 100 nm Ag film was deposited thermally through a patterned mask, leaving an active area of 10 mm².

2.3 | Characterization

Contact angle was measured by a Rame-Hart goniometer using sessile drop method. Atomic force microscopy (AFM) images were recorded using a multimode atomic force microscope (Digital Instruments, Nanoscope III) with tapping mode. X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) measurements were carried out using a PHI 5000 Versa Probe apparatus equipped with an Al Kα X-ray source (1486.6 eV). UPS was measured to acquire the work function with UV light source having He I emission at 21.2 eV, ca. 50W, with a take-off angle of 90°. Work function measurement was additionally done using Riken photoelectron spectrometer AC2. The cross-sectional and surface morphologies of films were recorded using Ultra plus (a member of Carl Zeiss) Field-Emission Scanning Electron Microscope (FESEM) at an accelerating operating voltage of 10 keV. The X-ray diffraction (XRD) was recorded using a Bruker D8 Advance with LYNXEYE detector and Cu absorber. The PSCs were measured under 1 sun irradiation using a Newport 91192A source meter with an AM 1.5G light spectrum at an intensity of 100 mW cm⁻². A mono silicon reference cell (PV Measurements, Inc.) was used to calibrate the 100 mW cm⁻² light intensity. The device performance was analyzed on an Agilent 4156C Precision Semiconductor Parameter Analyzer. The EQE spectra (DC mode) were obtained under ambient conditions without encapsulation using a 75-W Xe lamp (Enlitech, QE-R3011) light source. The steady state and time-resolved photoluminescence spectra were recorded using FL920 (FLSP920 Edinburgh Photonics model) fluorimeter with wavelength range from 700 to 800 nm.

### TABLE 1 Dipole moment values for different SAM molecules (irrespective of the binding surface), contact angles values and surface roughness for pristine NiOₓ and modified NiOₓ

| Surface      | Dipole moment [µ] | CA [deg.] | Roughness [nm] |
|--------------|-------------------|-----------|---------------|
| NiOₓ         | –                 | 18        | 1.55 nm       |
| PPA-NiOₓ     | 0.66              | 46        | 0.84 nm       |
| MPPA-NiOₓ    | 1.85              | 41        | 1.51 nm       |
| CNPPA-NiOₓ   | 3.59              | 42        | 0.93 nm       |

As a qualitative characterization of SAM formation on NiOₓ, CA measurement and roughness measurement using AFM were carried out on the pristine and the modified NiOₓ surfaces. As shown in Figure 1 and Table 1, the hydrophility of the three modified surfaces were rather similar, but less so than the pristine NiOₓ surface. Also, the surface roughness (Rₘₚₐₓ) did not significantly change, as shown in Figure 1 and Table 1. Nevertheless, for all the SAM-modified NiOₓ substrate, the surface became smoother than pristine NiOₓ. These may or may not influence the morphology of the perovskite film to be coated on the surface, as will be discussed later. The UV-vis
absorption spectra were also measured for the monolayer-modified NiO\textsubscript{x} films (shown in Figure S1). The SAM has little contribution to the UV absorption of NiO\textsubscript{x}.

To further confirm the formation of SAM on NiO\textsubscript{x} surface, XPS measurement was performed on the modified surface. The peaks associated with Ni2p\textsubscript{3/2}, O1s, P2p and C1s are shown in Figure 2 and tabulated in Table 2. The spectra for Ni2p\textsubscript{3/2} and O1s are deconvoluted and the results are shown in Figure S2. The Ni2p\textsubscript{3/2} region has two peaks, one at 853.9 eV, corresponding to the electronic state of Ni\textsuperscript{2+} and another peak at 855.4 eV corresponding to the electronic state of Ni\textsuperscript{3+} (deconvoluted in Figure S2).
Table 2 XPS peak values of bare and modified NiOx

| Substrate    | Ni2p [eV] | O1s [eV] | P2p [eV] | P2p Peak area [a.u.] | C1s [eV] |
|--------------|-----------|----------|----------|----------------------|----------|
| NiOx         | 861.0     | 854.3    | 529.2    | –                    | –        |
| PPA-NiOx     | 861.0     | 854.3    | 528.8    | 132.0                | 420.5    |
| MPPA-NiOx    | 861.0     | 854.3    | 528.6    | 132.1                | 396      |
| CNPPA-NiOx   | 861.2     | 854.7    | 529.0    | 132.0                | 328.5    |

Peak intensity for P2p peak to compare the surface coverage.

under the peak at 854.3 eV). The Ni2p3/2 and O1s peaks for all the modifiers are compared with that of the pristine NiOx. For PPA- and MPPA-modified surfaces, there is an insignificant shift of the peak position, indicating little change in local environment for NiOx, presumably because of the weaker electron-donating effect of H and MeO on the electron-density of the phenyl ring. For CNPPA-modification, a peak shift of 0.40 eV towards higher binding energy (B.E.) is observed, indicating a decrease in electron density, presumably because of the strong electron-withdrawing nature of the cyano group. These changes indicate local environment alterations near the NiOx surface. The peak at 861 eV indicates satellite peak associated with the main peak, occurring due to a sudden change in the Coulomb potential when the electron passes through the valence band.

The lower B.E. peak at ~529 eV in the O1s (Figure 2B) spectra for all conditions correspond to the lattice oxygen from the underlying NiOx. In Figure S2, the deconvoluted peaks, around ~530.2 eV is assigned to Ni2O3 and ~531.2 eV is assigned to NiOOH. The O1s peak shows shifts in case of modified NiOx as compared to pristine NiOx. With PPA modifier as the reference, there is a relative shift of 0.2 eV towards lower B.E. in the O1s peak for MPPA. This lower B.E. shift suggests a higher electron density which is in accordance with the smaller WF of NiOx after modification by MPPA.

The structure of the device used in the PSC is shown in Figure 3A. One of the major challenges of enhancing the charge flow in a PSC is to optimize the energy level alignment. In this study, the WF of the hole-transporting nickel oxide layer was modified using SAMs (inset Figure 3A) in order to elucidate its effect on the device performance. To analyze the energy levels after SAM modification, UPS measurements of pristine and modified NiOx were taken. The WF values, deduced from the low kinetic energy cut-off region (Figure S3A) and the Fermi cut-off region (Figure S3B) for pristine NiOx, PPA-, MPPA-, and CNPPA-modified NiOx, are 5.27, 5.23, 5.17, and 5.39 eV, respectively. Similar trend in WF was obtained using photoelectron spectroscopy in air (PESA) (Figure S4 and Table S1). Thus, the PPA SAM slightly decreased the WF, whereas the MPPA SAM further decreased the WF, presumably owing to the stronger electron-donating property of methoxy substituent. In contrast, the CNPPA SAM increased the WF value due to the electron-withdrawing substituent. The highest occupied molecular orbital of the standard MAPbI3 lies at -5.4 eV. Therefore, the modification of NiOx by CNPPA SAM brings the WF of NiOx closest to the valence band of MAPbI3. The energy level alignment for different layers along with different modifiers is shown in Figure 3A.

After the SAM modification, the perovskite layer was prepared by spin-coating the precursor solution onto the pristine and differently modified NiOx surfaces and annealing. The SEM images of perovskite film surface on different substrates (shown in Figure 4) exhibit meager difference in film coverage.

The grain size distribution and average grain sizes are compared (shown in Figure S5 and Table S2). No significant difference in the average grain sizes (~173-190 nm) was found. Further, the calculated full-width-at-half-maximum (FWHM) of the (110) peak from the XRD pattern of perovskite films (Figure S6 and Table S2) shows a similar trend in that smaller FWHM indicates better
FIGURE 3  (A) Schematic diagram of nickel oxide-based PSC (inset). Different para-substituted phosphonic acids used to modify the NiO$_x$ layer are phenyl phosphonic acid, 4-methoxyphenyl phosphonic acid, and 4-cyanophenyl phosphonic acid, with the net dipole direction indicated by the arrows. (B) Energy level alignment of perovskite film with pristine NiO$_x$ and differently modified NiO$_x$ (plot not to scale)

FIGURE 4  Scanning electron microscopy (SEM) of perovskite film surface on (A) pristine NiO$_x$, (B) PPA-modified NiO$_x$, (C) MPPA-modified NiO$_x$, and (D) CNPPA-modified NiO$_x$
crystallinity and larger grain size and vice versa. The cross-sectional images of perovskite films on pristine and modified NiO\textsubscript{x} surfaces (Figure S7) show a thickness of approximately 310 nm in all the cases (Table S2). Hence, it can be inferred that the SAM modification did not alter the perovskite film formation much, possibly due to similar wetting property of the surfaces. From these measurements, it is suggested that in correlating the changes in performance, the factors of perovskite thickness, coverage, or crystallinity may not be that critical. Additionally, the thickness for NiO\textsubscript{x} and PCBM in all cases is \textasciitilde60 and \textasciitilde70 nm, respectively.

To examine the effect of different SAM-modifications on the device performance, NiO\textsubscript{x} layer with and without SAM modification were used in fabricating devices with the structure: ITO/NiO\textsubscript{x}/SAM/CH\textsubscript{3}NH\textsubscript{3}PbI\textsubscript{3}/PCBM/Ag (Figure 3A). SAM modification changes the band edge of NiO\textsubscript{x}, which not only modulates the built-in voltage, but also other parameters.\cite{48} Figure 5A and Table 3 show the J-V curves and performance characteristics of the champion cell for each condition.

The champion cell based on pristine NiO\textsubscript{x} has a PCE of 17.02\% with an open circuit voltage (\textit{V}_{oc}) of 1.02 V, short-circuit current (\textit{J}_{sc}) of 22.98 mA cm\textsuperscript{-2} and a fill factor (FF) of 72.61\%. For the PPA-modified device, the \textit{V}_{oc} remained nearly the same as the pristine NiO\textsubscript{x} and is in agreement with the nearly unchanged work function of the nickel oxide after modification by PPA (Figure 3B). For the MPPA modification, the \textit{V}_{oc} slightly decreased from 1.02 to 0.99 V, while for the CNPPA modification, the \textit{V}_{oc} increased from

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**TABLE 3** Device parameters of PSCs fabricated on bare and modified NiO\textsubscript{x} HTL

| Device based on | \textit{V}_{oc} [V] | \textit{J}_{sc} [mA cm\textsuperscript{-2}] | FF [%] | PCE [%] |
|-----------------|-------------------|----------------|---------|---------|
| NiO\textsubscript{x} | Best | 1.02 | 22.98 | 72.61 | 17.02 |
| Avg±SD | 1.01±0.01 | 21.53±0.65 | 75.02±1.41 | 16.42±0.28 |
| PPA-NiO\textsubscript{x} | Best | 1.02 | 21.14 | 71.18 | 15.35 |
| Avg±SD | 1.01±0.01 | 20.99±0.74 | 69.17±3.09 | 14.76±0.31 |
| MPPA-NiO\textsubscript{x} | Best | 0.99 | 17.25 | 70.73 | 12.08 |
| Avg±SD | 0.98±0.01 | 16.81±1.46 | 68.71±4.35 | 11.29±0.71 |
| CNPPA-NiO\textsubscript{x} | Best | 1.06 | 23.48 | 73.96±1.83 | 18.15±0.21 |
| Avg±SD | 1.05±0.01 | 23.38±0.63 | 73.74±1.93 | 18.18±0.21 |
1.02 to 1.06 V. The CNPPA-modified NiOx has a WF closer to the valence band maximum (VBM) of MAPbI3 and MPPA-modified NiOx has a WF farther from VBM level of MAPbI3. The closer WF of CNPPA-modification provides a better built-in voltage, which is proportional to the difference in WF of the HTL and ETL.[16] The PPA- and MPPA-modified devices had decreased J_sc. Since the perovskite film has similar morphology and crystallinity, we attributed the decrease in J_sc to the dipole direction of these molecule pointing away from the NiOx surface, thus inhibiting the flow of holes towards NiOx. The CNPPA-modified device had increased J_sc, presumably due to a dipole directed towards the NiOx and thus facilitated the flow of holes to the NiOx surface.[49] The average fill factor was also improved with CNPPA-modification, indicating better interface and efficient charge transfer.[39,40] Overall, the observed trend in PCE was: MPPA-modified NiOx (12.08%) < PPA-modified NiOx (15.35%) < pristine NiOx (17.02) < CNPPA-modified NiOx (18.45%). As is clear from the analogous thickness of perovskite film on pristine and modified NiOx, the major contribution in the device performance arises from the band level alignment and dipole direction of the molecules. Better band alignment resulted in an increased V oc and the appropriate dipole direction resulted in an improved J_sc and thus enhanced device performance.[49] The corresponding external quantum efficiency (EQE) spectra are shown in Figure 5B, along with the integrated J_sc (values tabulated in Table S3), depicting a similar trend as that observed in the PCE. The enhanced EQE and J_sc in the case of CNPPA-modified NiOx based device indicates improved charge transfer at the interface of NiOx and perovskite layer.[16]

To further investigate the photo-carrier dynamics and observed efficiency trend due to modification of NiOx by SAM in PSC, steady state photoluminescence (PL) spectra of perovskite films on pristine and modified NiOx were measured, with the excitation wavelength of 470 nm and the range of detection from 700 to 850 nm. As shown in Figure 5C and Table S3, a clear difference in PL intensity was observed depending on the different underlayers on which the perovskite film was deposited. In case of perovskite layer alone without NiOx layer underneath, the PL intensity is the highest (shown in Figure S8). With NiOx HTL and SAM-modified NiOx inserted underneath the perovskite layer, various extent of quenching of the PL occurred. A maximum quenching of ~95% was observed in film incorporating CNPPA-modified NiOx. The quenching is suggested to be due to hole-charge transport from the perovskite layer to the NiOx layer so that radiative relaxation of the excitons to the ground state was diminished. On the other hand, PL quenching for MPPA-modified NiOx is the smallest (~83%), which means there is least charge transport from perovskite layer to NiOx layer. The quenching increased in the order of MPPA-NiOx < NiOx ~ PPA-NiOx < CNPPA-NiOx, roughly parallel to the trend in J_sc. To further elucidate the trend in PL quenching, we performed time-resolved photoluminescence (TRPL) measurement for the perovskite films on various NiOx substrates. The TRPL measurement was conducted using an excitor wavelength of 470 nm. The time-resolved PL decay plots (dotted plot in Figure 5D) were fitted using bi-exponential decay fitting (solid line in Figure 5D) and the parameters are tabulated in Table S4. The relatively faster decay components τ1 is attributed to charge carrier lifetime at the interface of perovskite and the transport layers, whereas the slower decay component, τ2, is related to lifetime in the bulk of the material. Average lifetime is calculated using the equation \[ \tau_{avg} = \frac{\sum_{i=1}^{2} R_i B_i}{\sum_{i=1}^{2} B_i} \] where \( R_i \) is the relative contribution of each lifetime \( R_i = B_i / \sum_{i=1}^{2} B_i \) in the bi-exponential decay model \( I(t) = A + B_1 e^{-t/\tau_1} + B_2 e^{-t/\tau_2} \) used to fit the TRPL spectrum. The observed quenching trend in PL and lifetime trend in TRPL correlate well with the J_sc.[15] Amongst all the conditions, CNPPA-modification enables the NiOx:Perovskite interface to have the fastest charge extraction as indicated by the shortest \( \tau_1 \), \( \tau_2 \) and \( \tau_{avg} \). The shortest lifetime for CNPPA-modification may indicate reduced trap states/better interfacial defect passivation, or superior interface leading to an increased fill factor. This reduction in trap states or interfacial defect passivation may result from binding of the localized negative charge of the cyano group with the positive Pb ions in perovskite, as suggested in previous report.[50]

Electrochemical Impedance Spectroscopy (EIS) was also used to evaluate the recombination in the devices. The EIS spectra were recorded in dark at open circuit voltage for each device. The Nyquist plot in Figure S9 shows that the device with CNPPA-modified NiOx exhibited the largest recombination resistance as compared to those devices with pristine or other modified NiOx. This indicates the reduction in recombination and efficient charge extraction and the trend observed was in agreement with our TRPL and PL quenching results. The device data statistics was estimated (results shown in box plot in Figure S10). The CNPPA-NiOx and pristine NiOx based devices show smaller distribution over the device performance, indicating better reproducibility. The dark storage stability of the devices was recorded. All the devices were stable and maintained similar performance for more than 50 days (Figure S11). The light soaking effect was also measured for all device conditions as shown in Figure S12. All devices showed stable PCE for above 1000 seconds. Hence, SAM modification didn’t have any adverse impact on the stability for the devices.
CONCLUSION

In conclusion, a systematic study of the effect of dipole-bearing SAM modification of the hole-transport layer of NiOx on the performance of inverted PSC was conducted. Molecules with different tail groups on the same phosphonic acid head group were used to modify the surface of nickel oxide layer. Phenylphosphonic acid used as the reference molecule did not significantly change the WF of nickel oxide layer. Modification by the acid carrying an electron-donating methoxy group resulted in a lower value of work function, owing to the positive dipole directing away from the oxide layer. The opposite effect was observed on NiOx modified with CNPPA, which increased the WF as a result of a negative dipole directing towards the oxide surface. The modification resulted in similar hydrophilicity of the surface and no major difference in film quality and thickness of the perovskite layer deposited on top. This simplifies the variables from various SAM modifications. A correlation between energy level alignment after SAM-modification and the open circuit voltage values can be seen. The best performing device is the CNPPA-modified NiOx-based device as CNPPA aligns the WF of NiOx closest to the valence band of perovskite, thus giving a larger Voc and enhancing the charge transport from perovskite to NiOx. The dipole direction may also contribute to the improved Jsc and the FF. More than 21% increase in the PCE was observed over the reference device based on pristine NiOx.

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