

Robust Ionic Anchoring at NiO_x Interfaces Enables Efficient Outdoor and Indoor Inverted Perovskite Solar Cells

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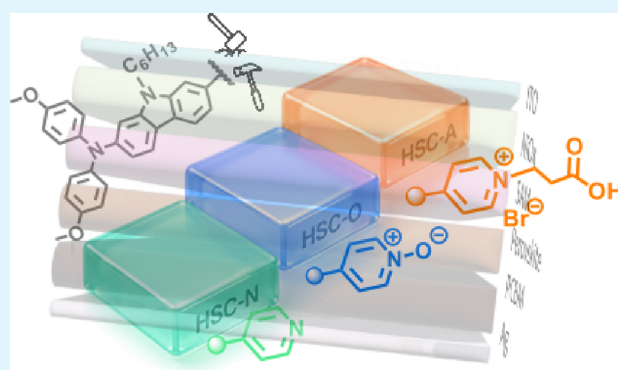
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ABSTRACT: The interfacial chemistry between hole-selective contacts (HSCs) and oxide substrates plays a critical role in governing charge extraction, interfacial recombination, and operational stability in inverted perovskite solar cells (PSCs). We designed three carbazole-based HSCs that share a D- π -A backbone but differ systematically in anchoring group chemistry and molecular ionicity: a neutral pyridine (HSC-N), a zwitterionic *N*-oxide (HSC-O), and an ionic pyridinium salt with a pendant carboxyethyl group and Br⁻ counterion (HSC-A), respectively. This molecular design isolates the anchoring-group effect and correlates it with interfacial energetics, dipole modulation, and device performance. Increasing molecular ionicity greatly enhances the dipole moment from 3.98 D for HSC-N to 17.11 D for HSC-A, effectively tuning the NiO_x surface energetics and promoting directional charge transport. After DMF/DMSO rinsing, HSC-A retained 72% of its initial surface loading, sufficient to deepen the valence band maximum at the NiO_x interface and most effectively suppress non-radiative recombination. Together, these effects yield champion PCEs of 21.42% and 19.52% for 1.57 eV and 1.68 eV perovskite absorbers, respectively. Under 1000-lux indoor illumination, HSC-A-based devices achieve an average PCE of 40.47% and retain nearly their initial efficiency after 34 days in ambient air. These results directly link anchoring-group design to device performance, highlighting ionic pyridinium-carboxylate contacts as an effective strategy for high-performance inverted PSCs.

KEYWORDS: hole-selective contacts, perovskite solar cells, indoor photovoltaics, donor-acceptor, anchoring group



1. INTRODUCTION

Organic-inorganic hybrid perovskite solar cells (PSCs) have emerged as a highly promising photovoltaic technology due to their profound gains in power conversion efficiency (PCE) and compatibility with low-cost solution processing.^{1–5} Inverted (p-i-n) architectures have attracted increasing attention owing to their negligible hysteresis, improved operational stability, and compatibility with tandem device configurations.^{6–8} Despite these remarkable advantages, the maximum efficiency of inverted PSCs is still predominantly constrained by the severe non-radiative recombination of charge carriers and charge transport barriers occurring at the device interfaces, which limit the open-circuit voltage (V_{OC}) and long-term operational stability of the devices. In particular, the interface between the hole transport layer (HTL), such as inorganic nickel oxide (NiO_x), and the perovskite film plays a crucial role in determination of overall device efficiency.^{9–11} Consequently, an effective interfacial treatment between the HTL and the perovskite layer is essential for mitigation of the buried interfacial losses.^{12–14} In recent years, the materials enabling formation of self-assembled monolayers (SAMs) have found widespread use as HSCs in inverted PSCs,

providing efficiency comparable to that of the hole transporting layers formed using conventional materials. Incorporation of hole-selective contacts (HSCs) enhances the device performance, primarily by facilitating efficient hole extraction and reducing non-radiative recombination losses at the perovskite interface.^{15–17} A key factor governing HSCs effectiveness is the rational design of anchoring groups, which play a pivotal role in determining interfacial binding strength, energy-level alignment, and long-term device stability. Robust anchoring not only ensures direct electronic coupling with the substrate but also suppresses interfacial degradation under operational stress.¹⁸ Over the past few years, the most efficient and widely studied SAMs have been those based on carbazole conjugate core, due to their excellent

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intrinsic hole transport mobility, high thermal stability and easily chemically modifiable conjugated framework.^{19–21} Despite these promising properties, the research on carbazole-based SAMs, particularly on those other than the ones anchored *via* phosphonic acid groups, remain limited. Anchoring groups capable of Lewis base interactions promote strong interfacial binding to oxide substrates (e.g., ITO or NiO_x), which is critical for efficient hole extraction, interfacial stability, and favorable energy-level alignment.^{22–24}

Recently, donor–acceptor (D– π –A) type SAMs have been shown to be effective HSCs for improving PSC performance mainly thanks to offering multifunctional sites, high dipole moment, and tunable optoelectronic and interfacial properties.^{25–27} Based on these properties, we designed and synthesized a series of carbazole-based D– π –A conjugated skeletons, systematically differing in their anchoring group chemistry, SAM-based HSCs, featuring asymmetric conjugated backbones. By strategically modulating the functional moieties within the molecular framework, we aimed to fine-tune the interfacial energetic alignment and passivate buried defects through specific chemical interactions. Specifically, we developed three interfacial modifiers for the treatment between the NiO_x HTL and the perovskite layer, i.e., a neutral pyridine-anchored molecule (HSC-N), a zwitterionic N-oxide derivative (HSC-O), and an ionic pyridinium salt bearing a carboxyethyl anchoring group (HSC-A). The main innovation of this study is the determination of anchor group effects by systematically modulating the ionicity in a fixed D– π –A system (Scheme 1). This structural motif induces large molecular dipole moments (from 3.98 D for HSC-N to a massive 17.11 D for HSC-A) that modulate interfacial energetics and promote directional charge transport. This platform enables a comprehensive investigation of the influence of anchoring groups on the physicochemical properties of HSCs and the corresponding PSC performance. Notably, the ionic HSC-A exhibits the strongest surface binding, retaining up to 72% of the initial surface loading even after aggressive solvent rinsing. Furthermore, HSC-A effectively increases the surface hydrophilicity and passivates deep-level traps, thereby significantly suppressing non-radiative recombination and prolonging bulk carrier lifetimes. As a result, inverted PSCs utilizing HSC-A as the interfacial treatment achieved an outstanding champion PCE results of 21.42% and 19.52% for perovskite absorbers with a bandgap 1.57-eV and

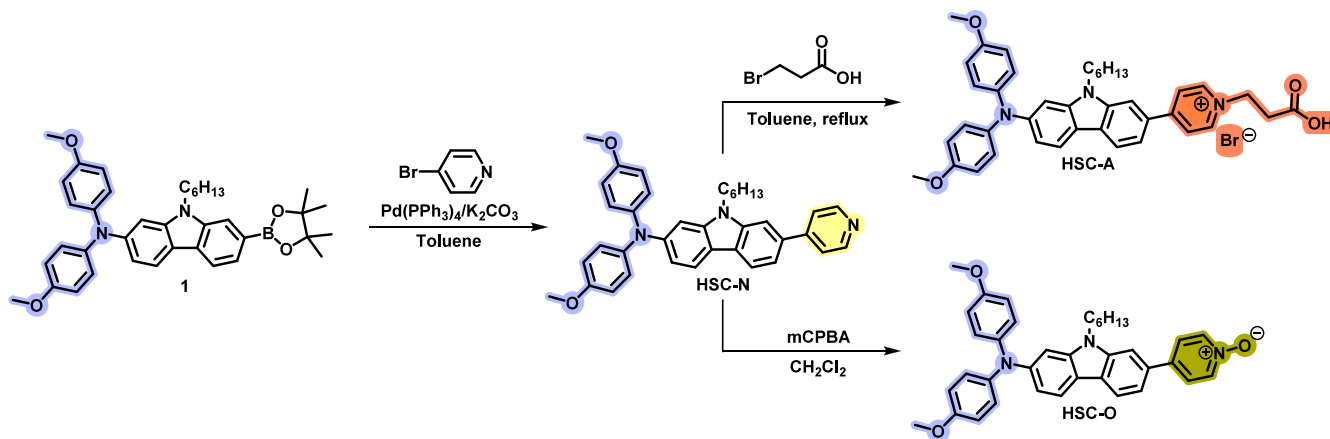
1.68-eV, respectively. Remarkably, under 1000-lux indoor WLED illumination, the wide-bandgap HSC-A devices delivered an average PCE of 40.47%, alongside exceptional long-term environmental stability.

2. RESULTS AND DISCUSSION

Carbazole, with its rigid structure, facile functionalization, and excellent hole-selective properties, is a versatile building block for optoelectronic materials and high-performance HSCs in inverted PSCs. Here, we designed D– π –A carbazole-based HSCs comprising a carbazole conjugated linker, bis(4-methoxyphenyl)amine donor, and pyridine acceptor unit, denoted as HSC-N in Scheme 1. HSC-N was synthesized *via* a palladium-catalyzed Suzuki coupling of boronate ester **1** with 4-bromopyridine. Subsequent *N*-alkylation of HSC-N with the corresponding 3-bromopropionic acid afforded HSC-A as a zwitterionic pyridinium salt bearing a carboxyethyl anchoring group and a bromide counterion. The zwitterionic derivative HSC-O, featuring an N-oxide acceptor, was obtained by oxidation of HSC-N with mCPBA. These carbazole-based HSCs were fully characterized by ¹H, ¹³C NMR spectroscopies, and high-resolution mass spectrometry. Detailed synthetic procedures and characterization data are provided in the Supporting Information and Figure S1–S9.

Density functional theory (DFT) calculations at the B3LYP/6-31G(d,p) level were performed to gain insight into the molecular geometry, energy levels, and orbital distributions of the HSCs. As illustrated in Figure 1a,b, the HSCs with D– π –A configurations exhibit HOMOs predominantly delocalized over the arylamine donor units and extending to the carbazole linker. In contrast, the LUMOs are mainly distributed over the carbazole backbone and the pyridine-based acceptor units. The partial spatial overlap between the HOMO and LUMO indicates pronounced intramolecular charge transfer (ICT) character, which is beneficial for hole extraction and transport. The calculated HOMO energy levels of HSC-N, HSC-O, and HSC-A are –4.62, –4.62, and –4.85 eV, respectively, while the corresponding LUMO levels are –1.20, –1.36, and –2.53 eV (Table 1). These results are consistent with the trends observed in the results of electrochemical measurements discussed below. DFT calculations (Figure 1c and Table 1) indicate that HSC-N, HSC-O, and HSC-A have dipole moments of 3.98, 7.16,

Scheme 1. Reaction pathways of Target HSCs



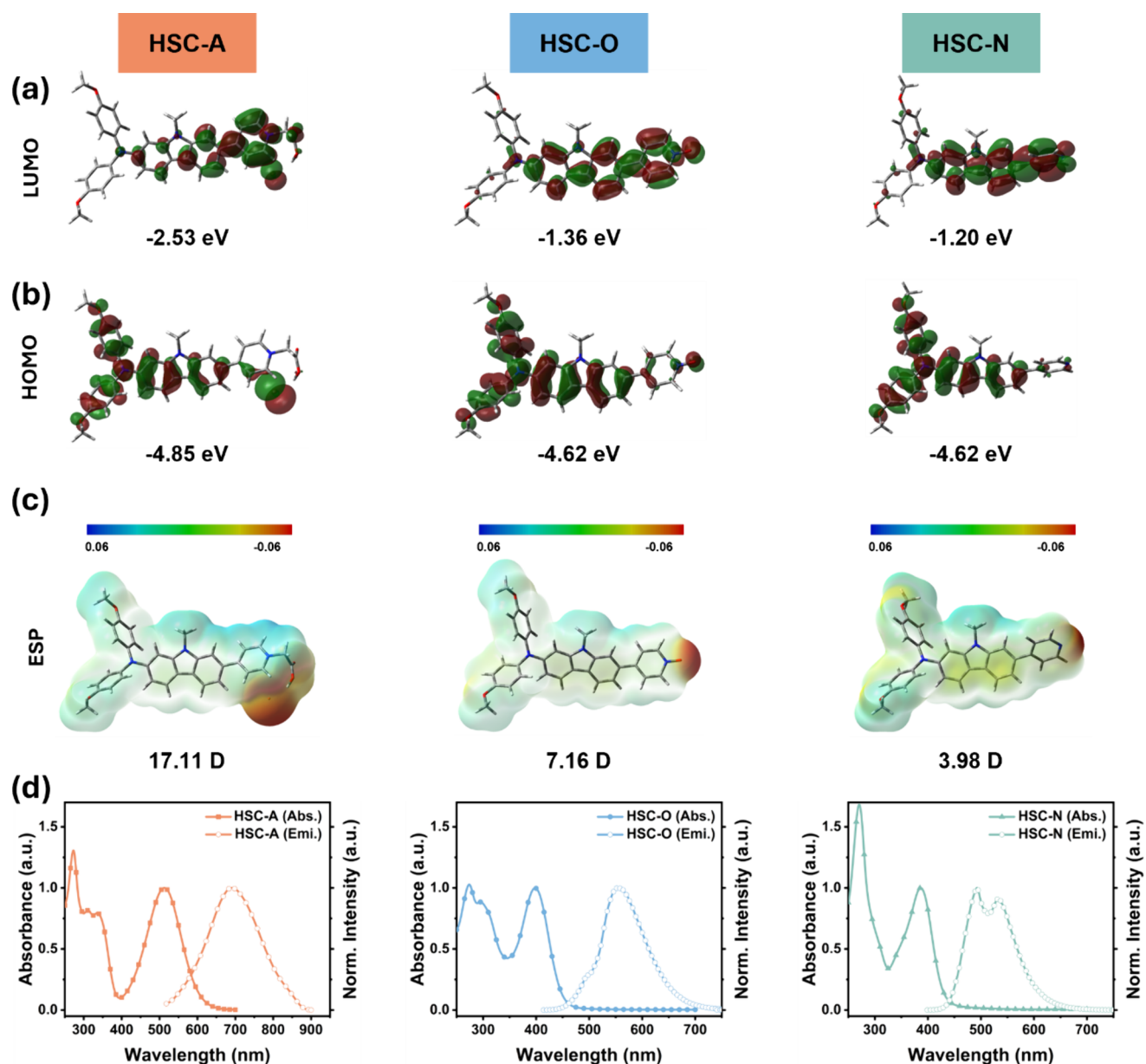


Figure 1. (a–c) DFT-derived frontier molecular orbitals and electrostatic surface potential (ESP) mapping of the HSCs. (d) Absorption and emission spectra of the HSCs.

Table 1. Photophysical, Electrochemical and Thermal Properties of the HSCs

HSC	HOMO ^a [eV]	LUMO ^a [eV]	dipole moment ^b [D]	HOMO ^c [eV]	LUMO ^d [eV]	$\lambda_{\max}$ ^e [V]	E_{0-0} ^f [V]	T_d ^g [°C]
HSC-N	-4.62	-1.20	3.98	-4.97	-2.16	387	2.81	353
HSC-O	-4.62	-1.36	7.16	-4.98	-2.30	398	2.68	327
HSC-A	-4.85	-2.53	17.11	-5.02	-2.84	513	2.18	218

^aby DFT calculations; ^bthe calculations were carried out using the Gaussian09 software package; ^cby DPV in DCM with ferrocene as an internal reference at 25 °C; ^dcalculated by ($E_{\text{HOMO}} + E_{0-0}$); ^emaxima absorption and emission peaks were obtained in chloroform; ^fthe value of E_{0-0} was determined from the intersection of absorption and emission spectra; and ^gby TGA.

and 17.11 D, respectively, suggesting that increasing molecular dipole moments may induce stronger interfacial polarization and more effectively modulate the electronic properties of the NiO_x/HSC interface, thereby facilitating hole extraction and charge collection.^{28–30}

To validate the calculations, the energy levels of the HSCs were experimentally determined by differential pulse voltammetry (DPV), and the optical bandgaps were estimated from the UV–vis absorption spectra. Representative spectra are shown in Figure S10, and the corresponding data are summarized in

Table 1. The HOMO levels of HSC-N, HSC-O, and HSC-A are -4.97 , -4.98 , and -5.02 eV, respectively. These values are higher than the valence band of the perovskite, facilitating efficient hole extraction. In contrast, the LUMO levels are -2.16 , -2.30 , and -2.84 eV for HSC-N, HSC-O, and HSC-A, respectively, indicating that the pyridinium–carboxylic acid anchoring group in HSC-A interacts stronger with the substrate than the pyridine or pyridine–oxide groups in HSC-N and HSC-O. The more positive LUMO levels of HSC-N and HSC-O relative to the perovskite conduction band, suggest that electron back transfer from the perovskite to the anode is effectively suppressed. The UV–vis absorption spectra of the HSCs were recorded in dichloromethane at a concentration of 1×10^{-5} M and are shown in Figure 1d. The corresponding data are collected in Table 1. The low-energy absorption maxima (λ_{max}) of the compounds were observed at 387 nm ($\epsilon = 3.67 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$), 398 nm ($\epsilon = 4.46 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$), 513 nm ($\epsilon = 3.35 \times 10^4 \text{ M}^{-1} \text{ cm}^{-1}$) for HSC-N, HSC-O and HSC-A, respectively. These bands are attributed to the intramolecular charge transfer (ICT) from the arylamine donor group to the pyridine acceptor, with delocalized π – π^* transition character. It is worth noting that the absorption maximum (λ_{max}) recorded for the HSC anchored through the non-conjugated carboxyethyl moiety is red-shifted relative to that of the one anchored through a system based on pyridinyl and pyridinyl oxide, indicating a stronger intramolecular charge transfer in the former. Thermal stability is a critical factor for the hole-selective materials in perovskite solar cells, as the latter are subjected to elevated temperatures under prolonged illumination. To evaluate these properties, the series of HSC compounds obtained were subjected to thermogravimetric analysis (TGA), and the corresponding curves are shown in Figure S11. All examined

materials exhibited high thermal stability, as their decomposition temperatures (T_d) exceeded 200°C . Notably, HSC-N and HSC-O show even higher T_d values above 300°C . These results indicate that the studied compounds possess sufficient thermal robustness to withstand long-term device operation without significant degradation.

To better mimic the conditions encountered during perovskite deposition, the HSC-modified substrates were rinsed with a DMF/DMSO mixed solvent to remove loosely bound species.³¹ Surface coverage was then quantified by desorbing the remaining HSC molecules using aqueous ammonia, followed by UV–vis analysis of the resulting solution. Molecular loading was subsequently calculated based on the Beer–Lambert law (Supporting Information Note 1 and Figure S12), enabling estimation of surface coverage after solvent rinsing. The change in the surface molecular loading after solvent rinsing depends strongly on the molecular structure. As shown in Figure 2a, HSC-A retains a significant portion of the initial loading (ca. 72%) after rinsing, indicating relatively strong anchoring to the nickel oxide surface. In contrast, HSC-O shows a more pronounced reduction in content, retaining only about one-third of its initial loading, suggesting the removal of a significant portion of weakly bound molecules upon washing. However, for HSC-N, the signal after rinsing falls below the detection limit, which makes it impossible to reliably determine the amount of material bound to the surface. The obtained results suggest that HSC-A forms a more robust and densely packed interfacial layer, while HSC-N, due to the anchoring group of lower polarity, most likely interacts much weaker with the modified surface. Interestingly, in the case of HSC-N, the surface nickel ions do not participate in any way in the interactions with the lone pair of pyridine electrons and formation of the Ni–N coordination

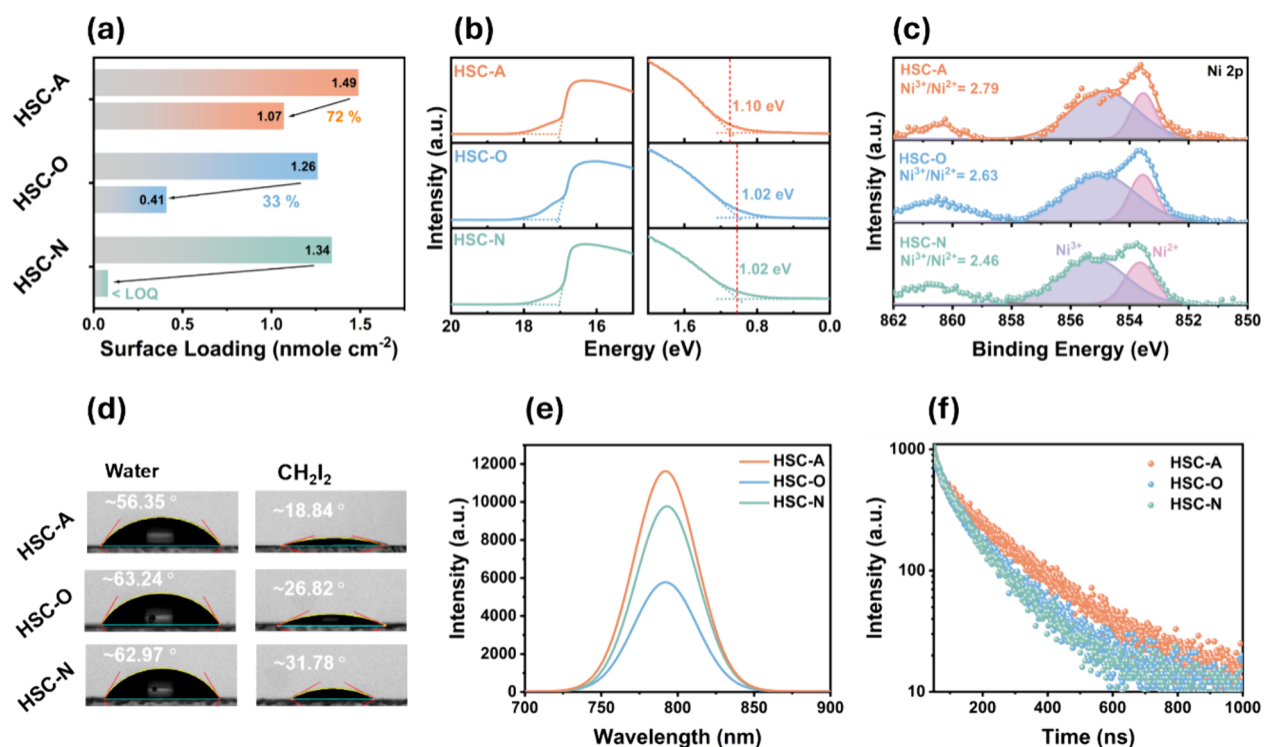


Figure 2. (a) Surface molecular loading of unrinsed HSC-modified substrates and after solvent rinsing. (b) UPS spectra. (c) XPS spectra of Ni 2p_{3/2} for HSC-modified NiO_x. (d) Contact angles of HSC-modified NiO_x surfaces. (e) PL spectra, (f) TRPL decay curves of perovskite films deposited on different substrates.

bond, a well-known fact from nickel chemistry, which suggests that sorption is determined solely by the possibility of hydrogen bonds formation.

To probe the interfacial energetics, ultraviolet photoelectron spectroscopy (UPS) measurements were carried out. The work function (ϕ) and valence band maximum (VBM) were determined from the secondary electron cutoff and onset regions using the eqs 1 and 2:

$$\phi = E_{\text{cutoff}} - 21.22 \text{ eV} \quad (1)$$

$$E_{\text{VBM}} = \phi - E_{\text{onset}} \quad (2)$$

As shown in Figure 2b, the VBM values of NiO_x modified with HSC-A, HSC-O, and HSC-N are -5.32 , -5.25 , and -5.24 eV, respectively (Table S1). This decreasing trend is consistent with the DFT calculations, which show that HSC-A exhibits a slightly deeper HOMO level (-4.85 eV) compared to those of HSC-O and HSC-N (-4.62 eV). The lower VBM of the HSC-A-modified interface suggests improved energy level alignment with that of the perovskite absorber, which is favorable for hole extraction and may help suppress interfacial recombination.

To further elucidate the interfacial interactions between the HSCs and the NiO_x surface, X-ray photoelectron spectroscopy (XPS) measurements were performed. As shown in Figure 2c and Table S1, the Ni $2p_{3/2}$ spectra can be deconvoluted into three main components. The peak at ~ 853 eV is assigned to Ni^{2+} species in the Ni–O coordination environment, while the feature at ~ 855 eV corresponds to Ni^{3+} species associated with Ni vacancies in oxygen-rich NiO_x . A satellite peak at ~ 860 eV is also observed, characteristic of the NiO_x lattice. The relative $\text{Ni}^{3+}/\text{Ni}^{2+}$ ratios are estimated to be 2.79, 2.63, and 2.46 for HSC-A, HSC-O, and HSC-N-modified surfaces, respectively. The variation in the $\text{Ni}^{3+}/\text{Ni}^{2+}$ ratio should be understood as an interfacial stabilization effect arising from the distinct anchoring chemistry of the HSCs. HSCs containing molecules with strong electron-withdrawing or coordinating groups (like pyridine or carboxylic acids) generally increase the Ni^{3+} concentration. This indicates enhanced oxidation of Ni^{2+} to Ni^{3+} , creating nickel vacancies and boosting the film's *p*-type conductivity.^{32,33} The carboxylic acid anchoring group in HSC-A can form stronger chemical interactions with surface Ni sites, while the ionic character induces a more pronounced interfacial dipole. These effects promote electron redistribution at the NiO_x/HSC interface, resulting in an increased proportion of surface Ni species exhibiting a Ni^{3+} -like electronic state in the XPS spectra. Since Ni^{3+} species are generally associated with nickel vacancies and enhanced *p*-type conductivity in NiO_x , the higher apparent $\text{Ni}^{3+}/\text{Ni}^{2+}$ ratio is consistent with improved hole extraction and the deeper valence-band maximum observed from UPS measurements. Together with the stronger interfacial binding, larger molecular dipole, and improved energy-level alignment, these results explain the superior interfacial charge transport and photovoltaic performance of HSC-A. Therefore, the transition from neutral (HSC-N) to zwitterionic (HSC-O) and finally to ionic (HSC-A) anchoring groups progressively enhances the coordinate binding strength and the interfacial electrostatic field. This strong chemical anchoring and polarization pull electron density from surface Ni atoms, shifting the Ni oxidation-state distribution toward

Ni^{3+} , which successfully induces a more pronounced *p*-type character favorable for hole transport.³⁴ Consistent with the XPS results, the *I*–*V* characteristics of $\text{ITO}/\text{NiO}_x/\text{HSC}/\text{Ag}$ devices show a similar trend in charge transport behavior (Figure S13). The slopes of the *I*–*V* curves follow the order of HSC-A > HSC-O > HSC-N, indicating that the HSC-A-modified interface exhibits improved charge transport capability, in agreement with its higher $\text{Ni}^{3+}/\text{Ni}^{2+}$ ratio and enhanced *p*-type characteristics.

The wettability and surface energy of NiO_x/HSC surfaces were evaluated to probe their macroscopic surface polarity. As shown in Figure 3d, the water contact angles for HSC-A, HSC-O, and HSC-N were measured to be 56° , 63° , and 62° , respectively, indicating that HSC-A has a relatively more hydrophilic surface. Notably, the reduced water contact angle observed for HSC-A can be attributed to both, the Br^- counterion and the positively charged pyridinium moiety, which enhances the surface polarity by increasing the density of hydrophilic ionic species at the NiO_x interface. A similar trend is observed for diiodomethane, with contact angles of 18° , 26° , and 31° for HSC-A, HSC-O, and HSC-N, respectively. Using water and diiodomethane as probe liquids, the total surface energies were calculated using the Wu model (Table S1).³⁵ The consistently lower contact angles of both polar and nonpolar probe liquids, collectively suggest a higher total surface energy and enhanced wettability, which are expected to promote uniform perovskite film coverage and support high-quality film formation.³⁶

The morphology of the perovskite films deposited on these substrates was then examined by the top-view SEM. All perovskite films studied exhibit dense and continuous morphologies without obvious pinholes (Figure S14).

We further investigated the bulk crystallization quality of the perovskite films using grazing-incidence wide-angle X-ray scattering (GIWAXS, see Supporting Information Note 2 and Figure S15). A distinct diffraction ring located at $q = 1 \text{ \AA}^{-1}$ corresponds to the perovskite (100) crystal plane. Compared with its counterparts, the perovskite film deposited on the HSC-A-modified surface exhibits stronger scattering intensity and sharper diffraction features, suggesting enhanced crystallinity and preferred crystal orientation. In contrast, the HSC-O-based sample shows a split peak in the corresponding 1D profile, which is likely associated with interfacial strain or defect-induced lattice distortion within the perovskite lattice.^{19–21} Steady-state photoluminescence (PL) and time-resolved PL measurements were performed to characterize carrier recombination and interfacial charge dynamics. As shown in Figure 2e, the perovskite film deposited on $\text{NiO}_x/\text{HSC-A}$ exhibits the highest PL intensity, indicating reduced non-radiative recombination at the $\text{NiO}_x/\text{perovskite}$ interface. This behavior suggests improved interfacial passivation, consistent with the stronger anchoring and higher surface coverage observed for HSC-A. In contrast, the $\text{NiO}_x/\text{HSC-O}$ sample shows a pronounced PL quenching, which may arise from more efficient interfacial hole extraction or additional quenching pathways associated with the polar N-oxide group.³⁴ The $\text{NiO}_x/\text{HSC-N}$ sample exhibits intermediate behavior. The TRPL profiles shown in Figure 2f were fitted using a bi-exponential function, yielding two characteristic components (Table S2). The fast component (τ_1), associated with interfacial charge transfer, is the shortest for HSC-O (2.13 ns), followed by HSC-N (2.72 ns) and HSC-A (3.15 ns), indicating more rapid charge extraction in

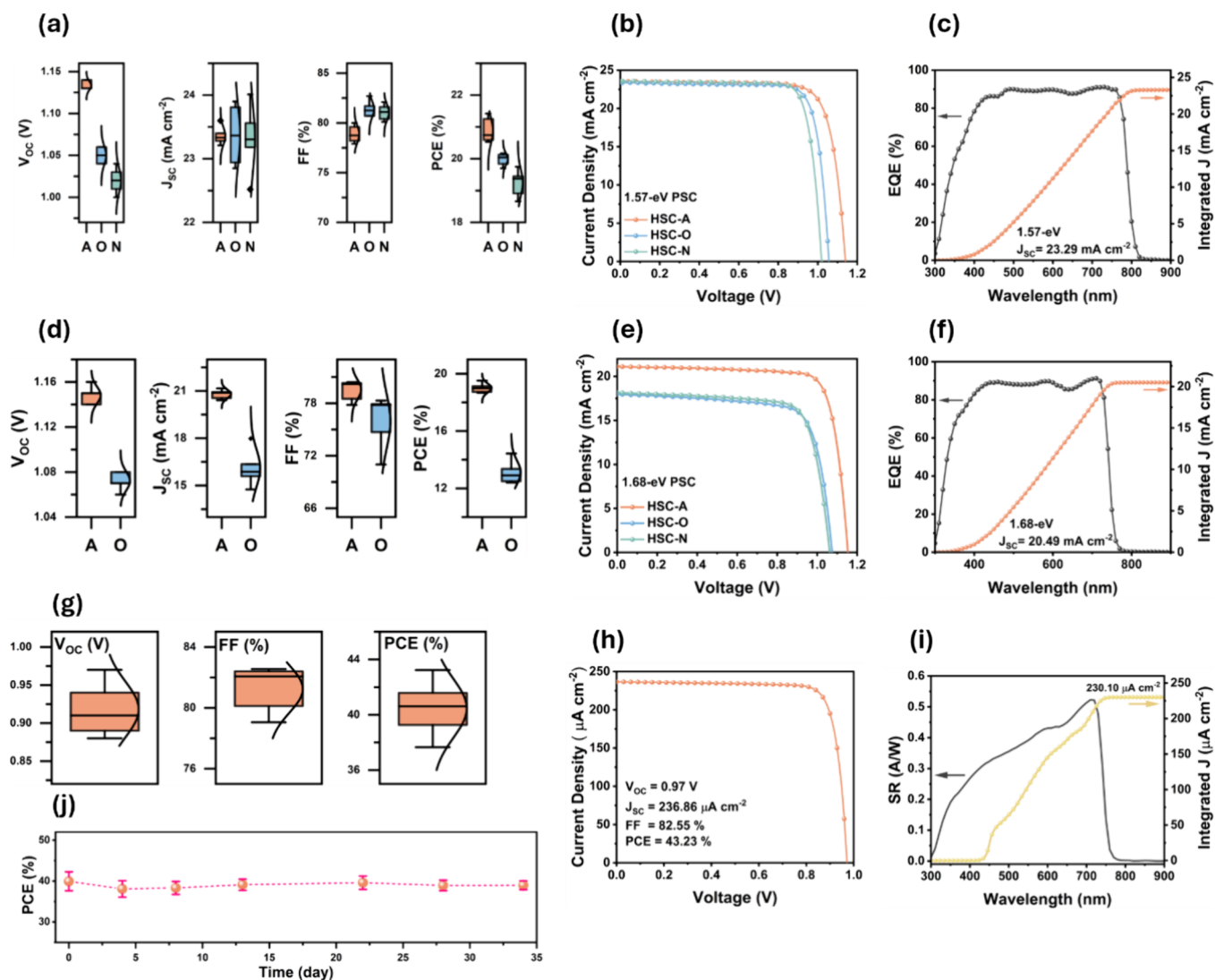


Figure 3. (a) Statistical distribution of photovoltaic parameters, (b) J - V curves of HSC-based 1.57-eV PSCs. (c) EQE spectrum and integrated J_{sc} curve of the 1.57-eV PSC. (d) Statistical distribution of photovoltaic parameters of 1.68-eV PSCs based on HSC-A and -O (e) J - V curves of HSC-based 1.68-eV PSCs. (f) EQE spectrum and integrated J_{sc} curve of the 1.68-eV PSC. (g) Statistical distribution of photovoltaic parameters of indoor photovoltaic devices. (h) J - V curve, (i) SR spectrum and integrated J_{sc} curve of the indoor photovoltaic device. (j) ISOS-D1 stability of unencapsulated devices.

HSC-O. In contrast, the slow component (τ_2), related to bulk recombination, is significantly extended for **HSC-A** (131.04 ns) compared to **HSC-O** (87.23 ns) and **HSC-N** (82.57 ns), suggesting suppressed recombination in the perovskite layer.³⁷ These results indicate a trade-off between charge extraction and recombination: although **HSC-O** facilitates faster interfacial charge transfer, **HSC-A** more effectively suppresses non-radiative recombination.

PSCs were fabricated with the device structure ITO/ NiO_x /HSCs/perovskite/PDADI/PCBM/BCP/Ag. In this device architecture, NiO_x serves as the hole transport layer, while the HSCs function as hole-selective interlayers at the NiO_x /perovskite interface. The distinct molecular dipoles and interfacial properties of these HSCs enable systematic investigation of how anchoring-group chemistry influences device physics and carrier dynamics, particularly interfacial energetics, charge extraction, and nonradiative recombination. Both 1.57 eV and 1.68 eV PSCs were

evaluated, with fabrication details provided in the [Experimental Section](#). For the 1.57-eV $\text{FA}_{0.85}\text{Cs}_{0.1}\text{MA}_{0.05}\text{PbI}_{2.95}\text{Br}_{0.05}$ perovskite the bandgap was estimated from the external quantum efficiency (EQE) spectrum (Figure 3c), and the corresponding photovoltaic performance is summarized in Figure 3a and Table S3, with the J - V curves shown in Figure 3b. From among the devices based on the three HSCs studies, those based on **HSC-A** deliver the highest performance (champion PCE of 21.42%), with an average PCE of $20.88 \pm 0.34\%$, along with an open-circuit voltage (V_{oc}) of 1.13 ± 0.01 V, a short-circuit current density (J_{sc}) of 23.36 ± 0.12 mA cm^{-2} , and a fill factor (FF) of $78.87 \pm 0.75\%$. The J_{sc} value agrees well with that integrated from the EQE spectrum (Figure 3c). In comparison, the **HSC-O**-based devices exhibit a PCE of $19.99 \pm 0.17\%$, with a V_{oc} of 1.05 ± 0.01 V and an FF of $81.38 \pm 0.71\%$, while the **HSC-N**-based devices show a lower PCE of $19.25 \pm 0.36\%$, with a V_{oc} of 1.02 ± 0.01 V and an FF of $81.07 \pm 0.68\%$. Notably, the

average J_{SC} values remain nearly identical for all devices, indicating that the differences in performance originate primarily from variations in V_{OC} and, to a lesser extent, FF . The significant increase in V_{OC} of HSC-A-based devices suggests a reduced non-radiative recombination at the NiO_x /perovskite interface. This improvement can be attributed to the stronger interfacial interaction introduced by the carboxyethyl anchoring group in HSC-A, which enhances interfacial contact and passivation. In addition, the larger dipole moment of HSC-A is likely to induce a more favorable interfacial energetics, facilitating hole extraction and further reducing the recombination losses.

For the 1.68-eV $Cs_{0.18}FA_{0.82}Pb(I_{0.8}Br_{0.2})_3$ perovskite, the device performances based on different HSCs are summarized in Figure 3e and Table S4. Statistical distributions for representative devices (HSC-A and HSC-O) are shown in Figure 3d. The HSC-A-based devices exhibit the best average performance, delivering an average PCE of $19.01 \pm 0.28\%$, with a V_{OC} of 1.15 ± 0.01 V, a J_{SC} of 20.81 ± 0.25 mA cm⁻², and an FF of $79.57 \pm 1.03\%$. The champion device achieves a PCE of 19.52%. The integrated current density obtained from the EQE is in good agreement with the J - V measurements (Figure 3f), confirming the reliability of the photovoltaic results. In comparison, the HSC-O-based devices show a significantly lower PCE of $13.12 \pm 0.73\%$, while the HSC-N-based devices yield a PCE of 14.19 ± 0.33 . The markedly improved performance of HSC-A is consistent with its superior interfacial properties, including stronger anchoring, favorable energetics, and suppressed recombination.

To further evaluate the applicability of HSC-A under practical low-light conditions, indoor photovoltaic performance of the devices based on the 1.68-eV architecture was investigated. As follows from the statistical data (Figure 3g and Table S5) and the corresponding J - V curves (Figure 3h), the HSC-A-based devices measured under WLED illumination at 1000 lux deliver an average PCE of $40.47 \pm 1.91\%$. The current density was further evaluated by integrating the spectral response (SR) of the device with the emission spectrum of the WLED source (Figure S16), yielding values consistent with the measured J - V results (Figure 3i). Finally, the stability of the devices was evaluated following the ISOS-D1 protocol. The unencapsulated HSC-A-based devices retained nearly their initial efficiency after 34 days of storage under ambient conditions (Figure 3j), indicating good environmental stability.

3. CONCLUSIONS

Three carbazole-based HSCs sharing the same D- π -A backbone but differing in anchoring chemistry were employed to elucidate the influence of interfacial binding strength and molecular ionicity on photovoltaic performance. Because the conjugated backbone remains unchanged, the observed differences can be attributed directly to the nature of the anchoring groups. Among the three materials studied, HSC-A exhibits the most favorable characteristics. Its ionic pyridinium-carboxyethyl functionality forms a robust interfacial layer that is more resistant to solvent rinsing than the neutral and zwitterionic counterparts. This stronger interfacial binding is accompanied by improved interfacial energetics and more uniform perovskite film formation. These results highlight the importance of anchoring group design in NiO_x -based inverted PSCs. The pyridinium-carboxyethyl motif provides an effective strategy to enhance interfacial stability, regulate energy level

alignment, and improve film quality. As a result, HSC-A reduces the recombination losses and improves interfacial contact, leading to a superior device performance with PCEs of 21.42% and 19.52% for 1.57-eV and 1.68 eV-PSCs, respectively. The applicability of HSC-A was further demonstrated in indoor photovoltaic devices based on the wide-bandgap architecture, which achieved an efficiency of up to 43.23% under WLED illumination. Overall, this study establishes a clear molecular design guideline for hole-selective contact materials, showing that anchoring group engineering plays a key role in governing buried interface energetics and defect passivation.

4. EXPERIMENTAL SECTION

4.1. Perovskite Thin Film Fabrication

For the 1.57-eV perovskite, the precursor solution was prepared by dissolving 228.76 mg of FAI, 677.67 mg of PbI_2 , 23.57 mg of CsCl, and 7.84 mg of MABr in a mixed 0.85 mL of DMF and 0.15 mL of DMSO. The precursor solution was spin-coated onto the substrate at 5000 rpm for 30 s. During the spin-coating process, 0.25 mL of chlorobenzene was dripped onto the spinning film 15 s before the end of the coating process. The resulting film was then annealed at 110 °C for 20 min to form the perovskite layer.

For the 1.68-eV perovskite, a 1.0 M precursor solution was prepared by dissolving 172 mg of FAI, 30 mg of CsCl, 110 mg of $PbBr_2$, and 354 mg PbI_2 in a mixed solvent of DMF and DMSO (4:1, v/v). The precursor solution was deposited via a two-step spin coating process at 1000 rpm for 10 s, followed by 5000 rpm for 30 s. During the second step, 0.15 mL of chlorobenzene was dripped onto the spinning film. The resulting films were annealed at 150 °C for 30 min to form the perovskite layer.

4.2. Device Fabrication of Perovskite Solar Cells

The perovskite solar cells were fabricated with the architecture of ITO/ NiO_x /HSC/perovskite/PDADI/ $PC_{61}BM$ /BCP/Ag. The ITO substrates were first sequentially cleaned by ultrasonication in detergent solution, deionized water, acetone, and isopropanol for 15 min each. After cleaning, the substrates were dried under a nitrogen flow and placed in a dry oven. Prior to use, the cleaned ITO substrates were treated with ultraviolet-ozone for 10 min. The NiO_x precursor solution was prepared by dissolving 55 mg of nickel acetylacetonate in 2.5 mL of ethanol with 1 vol % HCl as a stabilizing agent. The solution was spin-coated onto the ITO substrates at 5000 rpm for 30 s, followed by annealing at 250 °C for 30 min and a subsequent annealing step at 300 °C for 6 min in air. Subsequently, the HSC solution in ethanol (1 mg/mL) was spin-coated at 3000 rpm for 30 s and annealed at 100 °C for 10 min. The perovskite films were deposited following the same method described in the Perovskite Thin Film Fabrication section. For surface passivation, a PDADI solution (0.5 mg/mL in isopropyl alcohol) was spin-coated onto the perovskite layer at 5000 rpm for 30 s, followed by annealing at 100 °C for 5 min. Next, a $PC_{61}BM$ solution (20 mg/mL in chlorobenzene) was spin-coated at 1500 rpm for 30 s, followed by deposition of a BCP layer by spin-coating a 0.5 mg/mL solution in isopropyl alcohol at 6000 rpm for 10 s. Finally, a 100-nm-thick Ag electrode was thermally evaporated onto the BCP layer. The active device area was 10 mm², defined by the overlap of a 5-mm-wide patterned ITO bar and a 2-mm-wide Ag bar deposited via a metal mask.

■ ASSOCIATED CONTENT

Data Availability Statement

Data will be made available on request.

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsami.6c11066>.

Experimental details, including synthetic procedures, device fabrication conditions, NMR and mass spectrometry data, DPV spectra, TGA curves, GIWAXS patterns, TRPL measurements, surface energy analysis, and device J - V characteristics (DOCX)

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Z.-E.S.: designed the research and drafted the original manuscript. Y.-S.Y. and C.-P.C.: supervised the project. C.-H.C. and Y.-M.C.: synthesized the HSC-series compounds, characterized and performed the theoretical calculations. Z.-X.C.: and Y.-H.Z.: fabricated all the devices and conducted the major characterizations. I.K.: revised the manuscript and reviewed the data for accuracy. S.-C.J.: and J.-W.C.: conducted and analyzed the GIWAXS. Y.-R.L. and K.-M.L.: provided valuable discussions. All authors finalized the manuscript.

Notes

The authors declare no competing financial interest.

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REFERENCES

- (1) Kuo, C.-C.; Alhabshi, E.; Sung, C.-Y.; Lin, Y.-S.; Lin, Y.-C.; Qiu, W.-J.; McLachlan, M. A.; Lin, C.-T. Stress mitigation and defect passivation in CsPbI₂Br solar cells via controlled oxidation and thermal stress control. *J. Mater. Chem. A* **2025**, *13* (41), 35426–35434.
- (2) Das, C.; Kedia, M.; Tanko, K. T.; Wang, Y.; Njel, C.; Lira-Cantu, M.; Saliba, M. Tracing the Dynamic Chemical Transformations of Spiro-OMeTAD in Operating Perovskite Solar Cells. *Adv. Sci.* **2026**, No. e75107.
- (3) Tao, R.; Wang, G.; Li, Z.; Sun, N.; Bing, J.; Leung, T. L.; Angus, F. J.; Tang, J.; Liao, C.; Yi, J.; Bailey, C.; Liu, L.; Wang, Y.; Huang, G.; Lambert, A.; Yin, S.; Gong, B.; Cavallaro, A.-A.; Evans, D.; Griffith, M.; Kalantar-Zadeh, K.; Zheng, J.; Docampo, P.; McKenzie, D. R.; Mahmud, M. A.; Ding, K.; Ho-Baillie, A. W. Y. pH modulation for self-assembly-monolayer-type hole transport layer for efficient and stable perovskite-silicon double-junction solar cells. *Joule* **2026**, *10* (4), 102314.
- (4) Zhao, L.; Zhang, J.; Hua, Y.; Song, X.; Wu, C.; Chen, R.; Feng, Y.; Deng, H.; Su, J.; Gu, L.; Hui, W.; Chen, W.; Yang, C.; Song, L. Chemical bridging in a 2D/3D heterojunction via dual-anchoring functionalized molecules for efficient, stable and flexible perovskite solar cells. *Energy Environ. Sci.* **2026**, *19* (4), 1365–1373.
- (5) Zhang, J.; Zhao, L.; Su, Z.; Jia, W.; Deng, H.; Feng, Y.; Chen, R.; Su, J.; Wang, D.; Hua, Y.; Bao, Y.; Hui, W.; Gao, X.; Song, L. Disclosing Crystallization Kinetics for Efficient and Stable Perovskite Solar Cells. *Adv. Funct. Mater.* **2026**, *36* (6), No. e12025.
- (6) Han, Z.; Wang, Z.; Xia, Z.; Zhang, X.; Yang, J.; Liu, Y.; Zhai, Y.; Li, T.; Zhang, S.; Wang, L.; Jiang, Q.; You, J. Uniform phase distribution of wide bandgap perovskite for high-performance perovskite-silicon tandem solar cells. *Nat. Commun.* **2025**, *16* (1), No. 11470.
- (7) Zhuang, R.; Sun, A.; Tian, C.; Chen, C.; Liu, S.; Wu, X.; Han, S.; Cai, L.; Chen, J.; Chen, Q.; Zhao, Y.; Li, R.; Xue, T.; Cen, T.; Zhao, K.; Chen, C.-C. Co-deposited inverted perovskite photovoltaics towards 27% efficiency via vertical redistribution of self-assembled-molecules and in-situ crosslinking. *Nat. Commun.* **2026**, 5428.
- (8) Chen, Y.; Xu, C.; Liu, Z.; Xu, Y.; Lv, A.; Luo, M.; Wang, Z.; Jiang, D.; Mo, Y.; Li, H.; He, R.; Li, H.; Xia, R.; He, X.; Wang, Q.; Miao, D.; Gao, L.; Xie, J.; Yang, G.; Yu, D.; Zong, X.; Yang, Y.; Zhang, X.; Choy, W. C. H.; Yin, J.; Chen, Y.; Gao, J.; Chu, J.; Zhang, H. Stabilizing Efficient Perovskite/Silicon Tandem Solar Cells With Amorphous and Asymmetric Hole-Selective Contacts. *Adv. Mater.* **2026**, No. e73106.
- (9) Wang, D.; Liu, Z.; Qiao, Y.; Jiang, Z.; Zhu, P.; Zeng, J.; Peng, W.; Lian, Q.; Qu, G.; Xu, Y.; Zhang, Y.; Li, F.; Yan, L.; Wang, X.; Wang,

Y.-G.; Jen, A. K. Y.; Xu, B. Rigid molecules anchoring on NiOx enable >26% efficiency perovskite solar cells. *Joule* **2025**, 9 (3), No. 101815.

(10) Shi, Z.-E.; Hong, Y.-T.; Lu, C.-C.; Shih, P.-C.; Lu, S.-I.; Hsieh, H.-C.; Li, S.-R.; Lin, Y.-C.; Cai, Z.-X.; Lin, Y.-D.; Chen, C.-P. Isomeric halide engineering of self-assembled hole-selective layers for efficient indoor perovskite photovoltaics. *Chem. Eng. J.* **2026**, 527, No. 171787.

(11) Shi, Z.-E.; Fu, Y.-F.; Peng, B.-S.; Hung, T.-C.; Ganesan, D.; Balasaravanan, R.; Hong, S.-H.; Liu, C.-L.; Su, C.-J.; Chen, M.-C.; Chen, C.-P. Tailoring anchoring functionalities of self-assembled materials for efficient wide-bandgap perovskite solar cells. *Chem. Eng. J.* **2025**, 526, No. 171321.

(12) Li, K.; Zhang, Z.; Sun, S.; Dong, S.; Lu, X.; Liu, Y.; Dai, M.; Li, L.; Li, F.; Li, Z.; Zhang, H.; Zhai, Y.; Li, S.; Wang, H.; Zhu, X.; Chen, C.; Chen, J.; Zhu, T. Robust Adsorption of Self-Assembled Monolayer on NiO_x via Multiple Hydrogen Bonds for Stable Inverted Perovskite Solar Cells. *Adv. Mater.* **2026**, 38 (17), No. e72638.

(13) Shen, G.; Qiao, Z.; Zhao, X.; Li, Y.; Zhang, J.; Huang, Y.; Cai, Q.; Fang, L.; Li, X.; Mu, C. Tailored NiO_x mesoporous frameworks driving interface engineering towards durable and efficient inverted perovskite solar cells. *Nano Energy* **2026**, 152, No. 111896.

(14) Liu, Z.; Cai, X.; Song, Y.; Ge, H.; Wang, X.; Geng, C.; Zheng, Y.; Xu, H.; Wang, X.-F. A Reduction–Complexation Cascade Stabilizes the Buried NiO_x/Perovskite Interface for High-Performance Perovskite Solar Cells. *Adv. Funct. Mater.* **2026**, No. e75356.

(15) Tian, C.; Sun, A.; Chen, J.; Zhuang, R.; Chen, C.; Zheng, J.; Liu, S.; Du, J.; Chen, Q.; Cai, L.; Han, S.; Tian, F.; Chen, C.-C. Photostable donor–acceptor interface for minimizing energy loss in inverted perovskite solar cells. *Nat. Photonics* **2026**, 20 (3), 287–295.

(16) Li, F.; Liu, Y.; Huang, X.; Oh, C.-M.; Jiang, W.; Du, X.; Hwang, I.-W.; Jen, A. K. Y.; Park, S. H. Covalent Organic Framework-Templated Molecular Contacts for Inverted Perovskite Solar Cells. *ACS Energy Lett.* **2026**, 11 (4), 3377–3385.

(17) Yu, H.-W.; Liu, Y.-S.; Huang, H.-C.; Yu, M.-H.; Chen, Y.-H.; Lin, Y.-C.; Chang, Y.-H.; Lu, S.-I.; Lin, Y.-D.; Chueh, C.-C. Co-Assembly with Donor-Acceptor Self-Assembled Monolayers to Enhance Interfacial Hole Extraction, Wettability, and Crystallization in Inverted Perovskite Solar Cells. *Small* **2026**, 22 (23), No. e14211.

(18) Cen, T.; Zhuang, R.; Tian, C.; Sun, A.; Chen, Q.; Huang, R.; Zhao, Y.; Zhao, K.; Xue, T.; Li, R.; Chen, S.; Ni, H.; Zhao, T.; Chen, C.-C. Strengthening Substrate Anchoring of Polymeric Self-Assembled Monolayers for Efficient and Stable Inverted Perovskite Solar Cells. *Adv. Mater.* **2026**, 38 (17), No. e72622.

(19) Sun, A.; Tian, C.; Zhuang, R.; Chen, C.; Wu, X.; Han, S.; Cai, L.; Chen, Q.; Chen, J.; Du, J.; Chen, C.-C. Self-assembled monolayer with two-dimensional bounded network as robust bottom contact for inverted perovskite solar cells. *Joule* **2026**, 10 (6), No. 102380.

(20) Li, T.; Jiang, W.; Wang, T.; Yang, Y.; Liu, J.; Zhai, Y.; Liu, C.; Lin, M.; Tan, P.; Jiang, Z.; Wang, X.; Zhang, H.; Ni, Z.; Dai, Q.; Wong, C.-T.; Han, Z.; Wang, Z.; Zhang, X.; Jiang, Q.; Jen, A. K. Y.; Jiang, Q. Quasi-random oriented molecular contacts for inverted perovskite solar cells with improved efficiency. *Nat. Energy* **2026**, 11 (6), 866–875.

(21) Yang, J.; Qu, G.; Qiao, Y.; Cai, S.; Hu, J.; Geng, S.; Li, Y.; Jin, Y.; Shen, N.; Chen, S.; Jen, A. K. Y.; Xu, Z.-X. Flexibility meets rigidity: a self-assembled monolayer materials strategy for perovskite solar cells. *Nat. Commun.* **2025**, 16 (1), No. 6968.

(22) Puerto Galvis, C. E.; Sánchez, J. G.; Martínez-Ferrero, E.; Li, W.; Palomares, E. Dual functions of the anchoring groups in self-assembled molecules as effective hole-selective contacts in inverted perovskite solar cells. *EES Solar* **2026**, 774.

(23) Lee, G.-E.; Kim, J. H.; Kim, J.; Cho, H.-S.; Jung, S. M.; Hayashi, Y.; Ito, T.; Yamaguchi, K.; Saegusa, Y.; Takahashi, H.; Sato, H.; Miyasaka, T.; Kim, G. M.; Oh, S. Y. Interfacial engineering with a carbonyl-functionalized truxene self-assembled monolayer for high efficiency and stable NiO_x-based perovskite solar cells. *Chem. Eng. J.* **2026**, 537, No. 176106.

(24) Liu, Y.; Lin, X.; Zhao, X.; An, T.; Zhao, W.; Zhang, M.; Cui, L.; Weng, T.; Liu, B. Multi active sites regulation of buried interface defects

in n-i-p perovskite solar cells with tryptophan derivatives. *Chem. Eng. J.* **2026**, 534, No. 174910.

(25) Sheng, W.; Han, M.; Li, B.; Su, Z.; Liu, X.; Ghadari, R.; Sheng, J.; Shao, Z.; Cui, G.; Ding, Y.; Dai, S. Thiophene-Bridged Conjugated Self-Assembled Hole Transport Monolayer for Efficient and Stable Inverted Perovskite Solar Cells. *ChemSusChem* **2026**, 19 (1), No. e202502441.

(26) Wu, T.; Raju, T. B.; Li, H.; Shang, J.; Cao, Q.; Wu, L.; Narmandakh, K.; Song, J. T.; Staykov, A.; Zhang, C.; Ba, Q.; Zhao, L.; Nie, R.; Wang, P.; Yamada, S.; Dong, H.; Wang, Y.; Wang, S.; Matsushima, T.; Guo, Z. Hole-Selective Monolayer Molecules with Spatially Separated Carrier Orbitals and Twisted π -Skeleton for Inverted Perovskite Solar Cells and Modules. *ACS Nano* **2026**, 20 (6), 5318–5331.

(27) Afraj, S. N.; Velusamy, A.; Sharma, S.; Jacob, J.; Correa-Baena, J.-P.; Chen, M.-C.; Facchetti, A. Recent progress in molecularly tailored organic hole transporting materials for highly efficient perovskite solar cells. *J. Mater. Chem. C* **2025**, 13 (30), 15234–15289.

(28) Zhai, M.; Wu, T.; Du, K.; Chen, C.; Shibayama, N.; Narmandakh, K.; Khan, A. H. H.; Wang, Y.-C.; Shang, J.; Staykov, A.; Song, J. T.; Ida, S.; Wang, P.; Yamada, S.; Tamada, K.; Zhao, S.; Wang, A.; Guo, Z.; Matsushima, T.; Miyasaka, T.; Cheng, M. Interfacial dipole engineering by self-assembled molecules in n-i-p and p-i-n perovskite solar cells. *Nat. Commun.* **2026**, 17 (1), No. 2374.

(29) Jin, D.; Wang, X.; Zhou, J.; Liu, C.; Zhang, S.; Xu, J.; Sun, Z.; Liu, Q.; Peng, Z.; Gui, Y.; Shao, X.; Xin, H.; Jin, L.; Zhang, X.; Liu, R.; Ni, Z.; Yang, D.; Xue, J.; Wang, R. Unlocking Anchoring Group Mechanisms in Molecule-Based Hole-Selective Contacts for Perovskite Solar Cells. *Small Struct.* **2026**, 7 (1), No. e202500531.

(30) Ameen, S.; Lee, D.; Faheem, A. B.; Son, J. G.; Lee, Y.; Yoo, H.; Park, S.; Shin, Y. S.; Lee, J.; Seo, J.; Jang, H.; Roe, J.; Song, J. W.; Cho, S.; Park, Y.; Lee, K.-K.; Kim, J. Y.; Kim, D. S.; Kim, B. 3,6-Bis(methylthio)-9H-carbazole Based Self-Assembled Monolayer for Highly Efficient and Stable Inverted Perovskite Solar Cells. *Am. Ethnol.* **2025**, 137 (12), No. e202423206.

(31) Luo, Y.; Xue, J. Monolayer or not: Assembly configuration of SAMs in perovskite photovoltaics. *Joule* **2026**, 10 (4) 102390.

(32) Xu, Z.; Wang, J.; Liu, Z.; Xiao, C.; Wang, J.; Liu, N.; Zhang, J.; Huang, L.; Hu, Z.; Zhu, Y.; Liu, X. Self-Assembled Monolayer Suppresses Interfacial Reaction between NiO_x and Perovskite for Efficient and Stable Inverted Inorganic Perovskite Solar Cells. *ACS Appl. Mater. Interfaces* **2024**, 16 (40), 53811–53821.

(33) Liu, S.; Miao, T.; Zhang, Y.; Liu, Z.; Chen, W.; Park, N.-G. Progress in High-Efficiency Inverted Perovskite Solar Cells: Charge Transport, Interface Engineering, and Stability. *ACS Energy Lett.* **2026**, 11 (4), 3121–3147.

(34) Chen, C.; Lu, C.; Suo, Z.-Y.; Mu, X.; Yang, Y.; Cao, J. Stabilizing Ni³⁺-Rich NiO_x/Perovskite Interface via Dual Coordination for Efficient and Durable Perovskite Photovoltaics. *Adv. Mater.* **2026**, 38 (17), No. e21697.

(35) Wu, S. Polar and Nonpolar Interactions in Adhesion. *J. Adhes.* **1973**, 5, 39–55.

(36) Sangale, S. S.; Mann, D. S.; Lee, H.-J.; Kwon, S.-N.; Na, S.-I. Influence of interfacial roughness on slot-die coatings for scaling up high-performance perovskite solar cells. *Commun. Mater.* **2024**, 5 (1), No. 201.

(37) Hung, C.-M.; Shi, J.-H.; Tsai, H.-C.; Lin, C.-P.; Chen, B.-H.; Yang, S.-D.; Chou, P.-T. Interspersed Assembled Monolayers Enhance Hole Transport in High-Efficiency Organic and Perovskite Solar Cells. *J. Am. Chem. Soc.* **2025**, 147 (27), 23683–23695.