

Thermally Adaptive Interfacial Polymer Network Enables Space-Compatible Perovskite Solar Cells

Yiqiang Zhan

yqzhan@fudan.edu.cn

The State Key Laboratory of Photovoltaic Science and Technology, College of Future Information Technology, Fudan University <https://orcid.org/0000-0001-8391-2555>

Ran Wang

Fudan University

Liangliang Deng

Fudan University

Zhijie Hu

Fudan University

Xiaoguo Li

Jiangyin Jinghao New Energy Technology Co., Ltd.

Hongzhou Zhao

The State Key Laboratory of Photovoltaic Science and Technology, College of Future Information Technology, Fudan University

Dawei Zhao

The University of Manchester

Dan Li

The State Key Laboratory of Photovoltaic Science and Technology, College of Future Information Technology, Fudan University

Ligang Xu

Dalian Institute of Chemical Physics, Chinese Academy of Sciences

Qiang Guo

Fudan University

Tianyi Liu

The State Key Laboratory of Photovoltaic Science and Technology, College of Future Information Technology, Fudan University

Siqi Huang

Fudan University

Jiao Wang

Fudan University

Yingguo Yang

Fudan University <https://orcid.org/0000-0002-1749-2799>

Anran Yu

Fudan University

Article

Keywords:

Posted Date: September 2nd, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10813752/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.
[Read Full License](#)

Additional Declarations: There is **NO** Competing Interest.

Thermally Adaptive Interfacial Polymer Network Enables Space-Compatible Perovskite Solar Cells

Ran Wang¹, Liangliang Deng², Zhijie Hu¹, Xiaoguo Li², Hongzhou Zhao¹, Dawei Zhao³, Dan Li¹, Ligang Xu⁴, Qiang Guo¹, Tianyi Liu¹, Siqi Huang¹, Jiao Wang¹, Yingguo Yang⁵, Anran Yu^{1*}, Yiqiang Zhan^{1,6*}

¹The State Key Laboratory of Photovoltaic Science and Technology, College of Future Information Technology, Fudan University, Shanghai 200438, China

²Jiangyin Jinghao New Energy Technology Co., Ltd. Jiangyin, China

³Department of Materials, University of Manchester, Oxford Road, Manchester, M13 9PL, UK

⁴State Key Laboratory of Catalysis, Dalian National Laboratory for Clean Energy, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China

⁵School of Microelectronics, Fudan University, Shanghai 200433, China

⁶Institute of Optoelectronics, Fudan University, Shanghai 200438, China

Abstract

Metal halide perovskite solar cells (PSCs) are promising candidates for next-generation terrestrial and space photovoltaics owing to their high power-to-weight ratio, solution processability and intrinsic radiation tolerance. Yet their practical deployment, particularly in low-Earth-orbit (LEO) environments, is hindered by rapid temperature fluctuations and intense ultraviolet (UV) irradiation. These coupled thermomechanical stresses accelerate buried-interfacial degradation by promoting stress accumulation, defect generation and inefficient charge extraction at the interface between self-assembled monolayers (SAMs) and the perovskite absorber. Here we report an adaptive buried interface enabled by the glass transition of a thermoresponsive poly(2-hydroxyethyl methacrylate) (P2HM) interlayer inserted between the SAMs and the perovskite. The polymer undergoes *in-situ* structural adaptation near its glass-transition temperature ($T_g = 87.4^\circ\text{C}$), transforming into a compliant viscoelastic reticulated network that strengthens interfacial adhesion, bridges voids arising from incomplete SAM coverage and dynamically coordinates with undercoordinated Pb^{2+} sites. This adaptive interface not only passivates existing defects but also suppresses the continual generation of stress-induced interfacial defects during operation. The resulting PSCs achieve a power conversion efficiency (PCE) of 27.25% (certificated 27.19%) while retaining 90% of their initial efficiency after 540 rapid thermal cycles ($32^\circ\text{C min}^{-1}$ ramp rate) between -60°C and

+100°C and 93.4% after 316 h of intensified UV irradiation under 80 mW cm⁻². Our findings establish thermally adaptive buried interfaces as a general strategy for mitigating mechanically driven interfacial degradation, providing a design principle for durable perovskite photovoltaics operating under extreme terrestrial and space environments.

Introduction

Metal halide perovskite solar cells (PSCs) have rapidly emerged as one of the most promising photovoltaic technologies, with certified power conversion efficiencies (PCEs) now approaching 28%.^{1,2} Beyond high efficiency, their high power-to-weight ratio, low fabrication costs and intrinsic radiation tolerance make PSCs particularly attractive for applications where conventional photovoltaic technologies are limited, including lightweight terrestrial systems and low-Earth-orbit (LEO) spacecraft.^{3,4} However, practical deployment in these harsh environments requires perovskite devices to withstand rapid temperature fluctuations, intense ultraviolet (UV) exposure and extended high-temperature operation period to avoid degradation.⁵⁻⁷ In LEO, satellites routinely experience temperatures ranging from approximately -100°C to +100°C with the ramp rate of 10 °C min⁻¹ based on IEC 60068-2-14, substantially exceed those specified for terrestrial photovoltaic qualification standards.⁸⁻¹¹ These coupled thermal and mechanical stresses progressively degrade device integrity through stress accumulation, defect formation and interfacial failure, making stability under extreme environments one of the principal barriers to practical deployment of perovskite photovoltaics¹²⁻¹⁵

Although degradation occurs throughout the multilayer perovskite device, the buried interface between the self-assembled monolayers (SAMs) and the perovskite layer is particularly susceptible to thermomechanical failure. This interface governs perovskite nucleation, interfacial charge extraction and mechanical coupling, while simultaneously accommodating the mismatch in coefficients of thermal expansion (CTE). During film formation and repeated thermal cycling (TC), CTE mismatch generates residual stresses that distort the perovskite lattice, promote interfacial delamination and continuously generate new defect states, thereby accelerating non-radiative recombination and performance loss.¹⁶⁻²⁰ Preventing the continual accumulation of stress-induced defects at this buried interface is therefore essential for achieving durable PSCs capable of operating under realistic terrestrial and space conditions.

Considerable efforts have been devoted to mitigating thermomechanical degradation in perovskite photovoltaics. Dynamic materials, including self-healing fluoropolymer elastomers,²¹ flow-phase grain-boundary modifiers,²² photoisomerizable molecules and glass-transition-regulated additives,^{23,24} have demonstrated the ability to buffer strain and suppress mechanical damage within the bulk perovskite film or grain boundaries. Meanwhile, buried-interface engineering strategies based on molecular bridges,²⁵ dual-interface self-regulated interlayers,²⁶ CTE-gradient structures,²⁷ iodide-buffer layers,²⁸ cross-linked polymer networks²⁹ and various functional polymers have been applied to reduce interfacial recombination and improve operational stability.³⁰⁻³² Despite these advances, most existing interfaces remain structurally static after fabrication. Consequently, they cannot continuously adapt to evolving thermomechanical stresses generated during rapid temperature cycling, sustained high-temperature operation or intense UV exposure, where new interfacial defects are continuously produced throughout device operation. Developing adaptive buried interfaces capable of dynamically responding to extreme operating conditions therefore remains an outstanding challenge.³³

Here we report a glass-transition-enabled adaptive buried interface by introducing a poly(2-hydroxyethyl methacrylate) (P2HM) reticulated network between the SAMs and the perovskite layer. Owing to its glass-transition temperature ($T_g = 87.4^\circ\text{C}$), the polymer can be activated during annealing (100°C), and P2HM transforms into a compliant viscoelastic network with enhanced segmental mobility, strengthening interfacial adhesion, accommodating thermally induced strain and bridging interfacial voids originating from incomplete SAM coverage (Figure 1a, b). Simultaneously, the thermally reconstructed polymer exposes additional hydroxyl groups (-OH) that dynamically coordinate with undercoordinated Pb^{2+} ions, enabling sustained defect passivation while suppressing the continual generation of stress-induced interfacial defects during operation. As a result, the adaptive buried interface simultaneously improves crystallization, charge extraction and mechanical robustness, enabling PSCs with a PCE of 27.25%. More importantly, the optimized devices retain 90% of their initial efficiency after 540 rapid thermal cycles between -60 and $+100^\circ\text{C}$, 88.7% after 450 h of thermal aging at 100°C and 93.4% after 316 h of accelerated UV irradiation under 80 mW cm^{-2} . These findings establish adaptive buried interfaces as a general design strategy for mitigating thermomechanically driven interfacial degradation and provide a promising framework for durable perovskite photovoltaics operating in extreme terrestrial and space environments.

Results and Discussion

Mechanism for SAMs Interface Optimization

We first study the electrostatic potential (ESP) of the monomeric representation of the P2HM molecule, namely P2HM (Supplementary Figure 1). The electron-rich units of the carbonyl group (C=O) and -OH in P2HM exhibit negative ESP (red), which could effectively passivate Lewis acid defects (such as undercoordinated Pb^{2+}) by forming coordination covalent bonds.³⁴

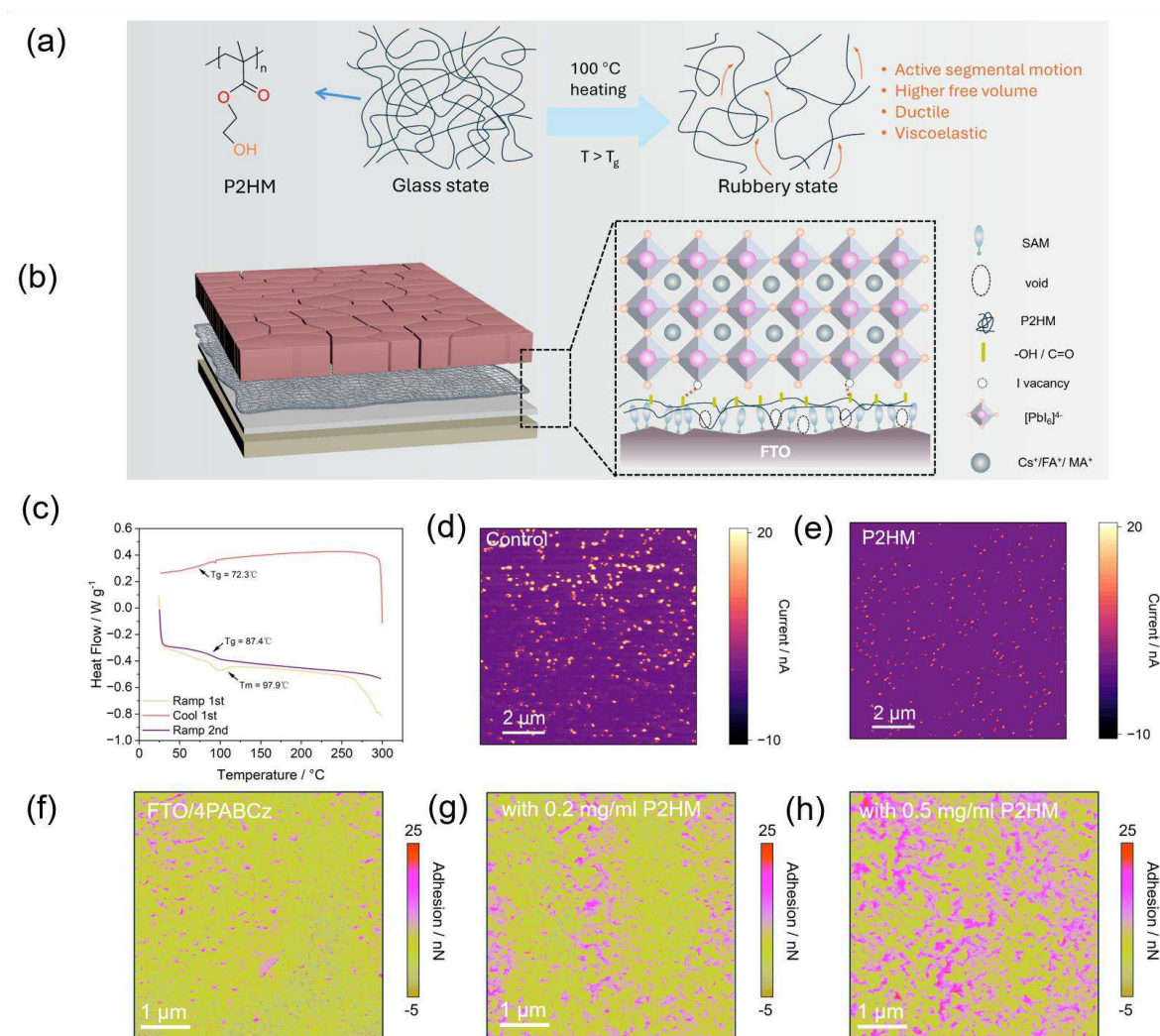


Figure 1 (a) Chemical structural formula of P2HM and a schematic diagram of segmental motion during the transition from the glassy state to the viscoelastic state. (b) Schematic diagram of the perovskite film after modification with P2HM. (c) DSC curves of P2HM ramp-cool cycle. C-AFM images of the FTO/4PABCz (d) and FTO/4PABCz/P2HM (e) films. (f-h) Adhesion AFM of different conditions films.

Differential scanning calorimetry (DSC) was used to analyse the dynamic transformation of P2HM during annealing, as shown in Figure 1c.³⁵ During the first ramping, P2HM exhibited an endothermic peak at approximately 98°C, which is attributed to enthalpy relaxation associated with the release of the polymer's thermal history and molecular rearrangement.³⁶ This process suggests that the initially frozen polymer chains undergo structural relaxation, accompanied by the disruption of local hydrogen-bonded aggregates and increased segmental mobility. As a result, previously buried -OH and C=O become more accessible, while the polymer evolves towards a more compliant and viscoelastic state.³⁷ This structural evolution is further supported by solid-state (SS) ¹H NMR (Supplementary Figure 2), heat treatment at 100°C triggers P2HM segment reconstruction, leading to differentiation of the local chemical environment of protons in the chains.³⁸ Simultaneously, changes in -OH groups related signals indicate a rearrangement of the hydrogen bond network within the P2HM. This thermally induced molecular reconstruction facilitates the exposure of more -OH and C=O, enhancing their potential for interaction with Pb²⁺ and interfacial defects in the perovskite, thereby achieving more effective dynamic interfacial passivation and stress relief. Upon the second heating cycle, only the characteristic *T_g* remains, confirming that the initial endothermic event originates from irreversible structural relaxation, whereas the subsequent glass transition corresponds to the reversible transformation between the glassy and viscoelastic states.^{39,40}

Conductive atomic force microscopy (C-AFM) results in Figure 1d, e show that the FTO substrate covered with SAMs 4-(9'-Phenyl-9H,9'H-[3,3'-bicarbazol]-9-yl) butyl)phosphonic acid (4PABCz) exhibits pronounced high-current regions (yellow color), whereas the FTO/4PABCz/P2HM substrate displays substantially smaller high-current areas, indicating more complete coverage of the underlying FTO surface. Consistently, the median current decreases from 0.88 nA for the 4PABCz substrate to 0.32 nA after P2HM deposition (Supplementary Figure 3), suggesting that the polymer interlayer effectively reduces exposed conductive pathways arising from incomplete SAM coverage and thereby suppresses leakage-current-induced non-radiative recombination.⁴¹ To further investigate the interfacial morphology, the adhesion AFM was performed on the FTO/4PABCz substrate and on samples coated with 0.2 and 0.5 mg/mL P2HM (Figure 1f-h). The FTO/4PABCz substrate exhibits isolated regions with relatively high adhesion, which are attributed to direct interactions between the exposed FTO surface and the AFM probe. Upon increasing the P2HM concentration, these high-adhesion domains gradually expand into discontinuous network pattern, indicating preferential localization of the polymer-rich.⁴² Simultaneously, peak force

quantitative nanomechanical mapping (PFQNM) acquired from the same areas (Supplementary Figure 4) reveals a corresponding evolution in the local mechanical response. This behavior suggests that P2HM initially accumulates at energetically favorable sites, such as exposed FTO regions, surface depressions and incompletely covered SAMs domains, before progressively bridging adjacent regions through chain entanglement and lateral spreading to form a mechanically compliant interconnected network⁴³

Enhancing Crystallisation via buried interface modification

To elucidate the chemical interaction between P2HM and perovskite, Fourier transform infrared (FTIR) spectroscopy was performed on PbI₂/P2HM mixtures after solvent removal (Figure 2a and Supplementary Figure 5). The characteristic C=O stretching vibration of P2HM shifts from 1726 to 1722.1 cm⁻¹ upon mixing with PbI₂, accompanied by the emergence of two additional bands at approximately 1659 and 1628.1 cm⁻¹ while the original C=O remains.⁴⁴ Additionally, -OH group shifts from 3407.6 to 3509.8 cm⁻¹. The shifts indicate that the C=O and -OH groups in P2HM experience different coordination environments. These results demonstrate the formation of coordination interactions between P2HM and PbI₂, which may contribute to defect passivation and interfacial stabilization in perovskite films.⁴⁵

To verify that such coordination is preserved at the buried interface of complete perovskite films, X-ray photoelectron spectroscopy (XPS) measurements were performed on peeled buried interfaces prepared using a UV-curable adhesive method.⁴⁶ As shown in Figure 2b, the control film displays two peaks at 143.17 and 138.27 eV corresponding to 4f_{5/2} and 4f_{7/2} of Pb 4f, respectively, which are shifted 0.20 eV in the P2HM-treated film, confirming a change in the electronic environment of Pb element arising from coordination with the polymer interlayer. Furthermore, crystallographic analysis by X-ray diffraction (XRD) confirmed that the P2HM-treated film exhibited a stronger (100) peak at 14° compared with the control film (Supplementary Figure 6a), together with reduced ratios of (100)/(110) peak and (100)/PbI₂ (~12.7°) peak, highlighting the improved crystallinity (Supplementary Figure 6b).⁴⁷ Two-dimensional grazing-incidence wide-angle X-ray scattering (GIWAXS) patterns with top and bottom perovskite sides of control film and P2HM-treated film (Figure 2c-d and Supplementary Figure 7) reveal similar diffraction features, accompanied by a weaker PbI₂ scattering ring in the P2HM-treated film.⁴⁸

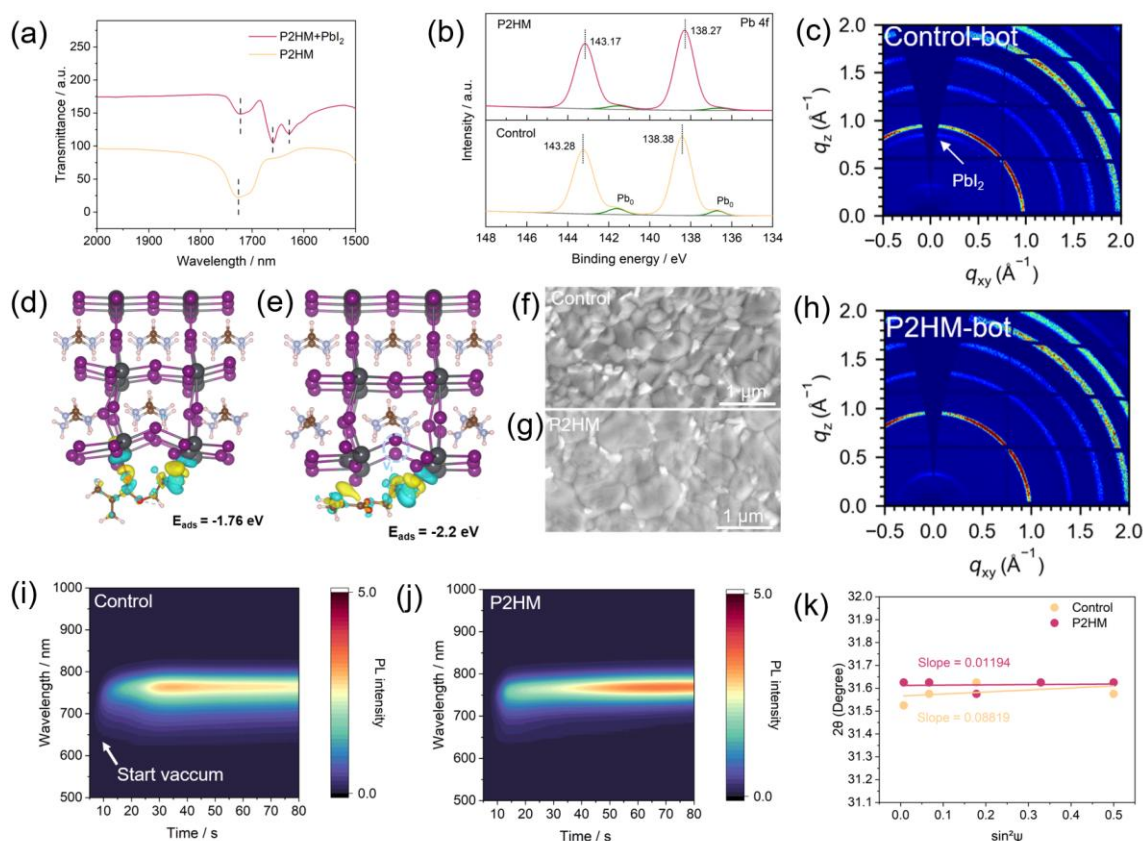


Figure 2 (a) FTIR spectra of P2HM and P2HM/PbI₂ mixture. (b) XPS spectra of bottom side perovskite films w/o and with P2HM-treated. (c, d) GIWAX patterns of the bottom side perovskite film before and after treatment at a grazing incidence angle of 0.2°. Simulations of (e) P2HM adsorbed on the perovskite slab and (f) with an I vacancy. (I, j) *In-situ* PL spectra for tracking control and P2HM-treated films during flash vacuum process. (k) Linear fit of $2\theta - \sin^2(\Psi)$ for the control and P2HM-treated films.

To elucidate the atomic-level origin of the interfacial coordination, density functional theory (DFT) calculations were performed using the HEMA monomer as a simplified structural model of P2HM on the FAPbI₃ surface. Iodine vacancy (V_I) defects are known to play a role in nonradiative recombination and ion migration, which reduces the efficiency of devices.⁴⁹ Therefore, our computational study focused on the interactions between both pristine and V_I defect-containing surfaces (Supplementary Figure 8 a, b). On the defect-free surface, the optimized HEMA molecule adopts a bidentate adsorption configuration, in which the C=O and -OH groups coordinate with neighbouring surface Pb²⁺ (Figure 2e and Supplementary Figure 8c), yielding a binding energy of -1.76 eV. The interaction becomes markedly stronger in the presence of a V_I surface, as illustrated in Figure 2f and Supplementary Figure 8d. HEMA undergoes a conformational rearrangement to adopt a more planar adsorption geometry,

allowing the -OH to interact more effectively with undercoordinated Pb^{2+} . This corresponds to a substantially increased binding energy of -2.20 eV, 0.44 eV lower than that on the pristine surface, indicating a pronounced thermodynamic preference for defect sites.⁵⁰ This strong molecular-perovskite interaction enhances surface anchoring strength, stabilizes undercoordinated Pb^{2+} and passivates surface V_I , all of which are expected to improve the performance and stability of the perovskite.

Top-view and peeled buried-surface scanning electron microscopy (SEM) images reveal that the P2HM-treated perovskite forms a denser and more homogeneous microstructure than the control film, accompanied by an increase in the average grain size from 576.2 to 679 nm (Figure 2g, h and Supplementary Figure 9,10). Cross-sectional SEM further shows continuous columnar grains extending across the film thickness, indicating improved vertical crystallization and reduced structural discontinuities (Supplementary Figure 11). AFM was used to observe the buried surface morphology of the film. As shown in Supplementary Figure 12, the root mean square roughness (RMS) of the P2HM-treated film was significantly lower than that of the control film (38.6 nm and 32.8 nm, respectively), consistent with the improved wetting behavior of the decreased contact angle (Supplementary Fig. 13).⁵¹ The combined observations demonstrate that the adaptive P2HM interlayer promotes the formation of a more compact and uniform buried interface.⁵²

To understand how the buried interface regulates crystallization, the film formation process was monitored by *in-situ* photoluminescence (PL) spectroscopy (Figure 2i, j). Compared with the control, the P2HM-treated film exhibits a delayed PL evolution, characterized by an initial decrease in band-edge emission followed by a gradual increase in intensity during crystal growth (Supplementary Figure 27). The maximum PL intensity is reached at 65 s, substantially later than that of the control film (31 s), indicating suppressed initial nucleation and prolonged crystal growth. Such delayed crystallization favours Ostwald ripening and the formation of larger grains with reduced defect density.⁵³ This behaviour is consistent with strengthened interfacial interactions between the precursor and the thermally activated P2HM layer, which increase the nucleation barrier and moderate crystallization kinetics.⁵⁴

Grazing Incidence X-Ray Diffraction (GIXRD) analysis revealed a significant shift of the diffraction peak to lower angles with increasing tilt angle (ϕ) for the control perovskite film, indicating considerable compressive stress (20.95 MPa) and lattice distortion (Figure 2k and Supplementary Figure 15). While the P2HM-treated perovskite film exhibited a mitigated peak

shift and a flatter slope in the corresponding 2θ - $\sin^2(\varphi)$ curve, demonstrating a substantial reduction in residual strain (2.84 MPa). The formulas for calculating the strain values are given in Equations (1),⁵⁵

$$\sigma = -\frac{E}{2(1+\nu)} \frac{\pi}{180^\circ} \cot \theta_0 \frac{\partial(2\theta)}{\partial(\sin^2 \psi)} \quad (1)$$

where E is the perovskite modulus (10 GPa) and ν is the Poisson's ratio of perovskite (0.3). This strain relief was attributed to the presence of the P2HM polymer network as a buffer at the buried interface, which can effectively accommodate the lattice mismatch.⁵⁶

Improved Carrier Dynamics and Reduced Non-Radiative Recombination

The influence of P2HM on the buried interfacial electronic structure was first examined by Kelvin probe force microscopy (KPFM). As shown in Figure 3a, b, the P2HM-treated interface exhibits a more homogeneous surface potential distribution than the pristine 4PABCz layer, accompanied by a slight increase in the contact potential difference (Supplementary Figure 12), indicating a more uniform interfacial energy landscape and enhanced p-type character (Supplementary Figure 16).⁵⁷

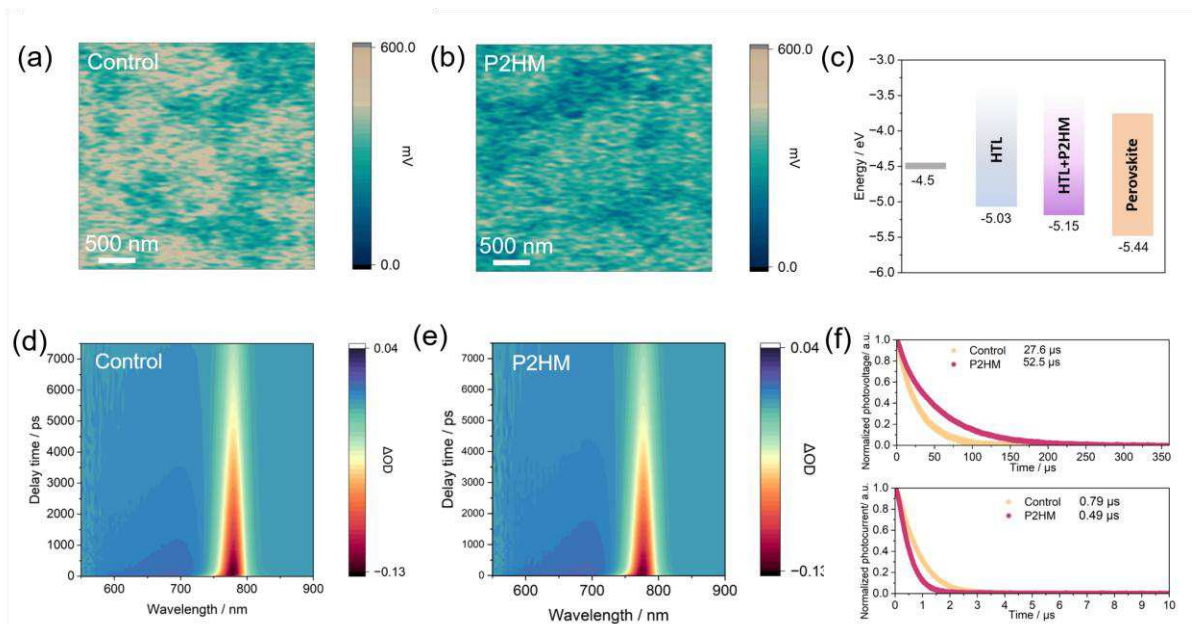


Figure 3 KPFM images of the (a) Control and (b) P2HM-treated film (bottom side) measured for same area of Supplementary Figure 12. (c) Energy-level diagrams of FTO/4PABCz, FTO/4PABCz/P2HM and perovskite films. (d) Pseudo-colour TA spectra of FTO/4PABCz/perovskite and FTO/4PABCz/P2HM/perovskite films. TPV (f) and TPC (g) decay curves of control and treated devices.

Ultraviolet photoelectron spectroscopy (UPS) further reveals that introducing P2HM shifts the highest occupied molecular orbital (HOMO) level of the 4PABCz from -5.03 to -5.15 eV, bringing it closer to the valence-band maximum of the perovskite (-5.44 eV) (Figure 3c and Supplementary Figure 17).⁵⁸ This more favorable energy-level alignment facilitates hole transfer across the buried interface while preserving the optical bandgap of the perovskite, as confirmed by ultraviolet-visible (UV-vis) spectroscopy (Supplementary Figure 18). The observed energy-level modulation is attributed to interfacial dipole redistribution arising from the hydroxyl-rich P2HM layer, which alters the local electronic environment of the 4PABCz surface.⁵⁹

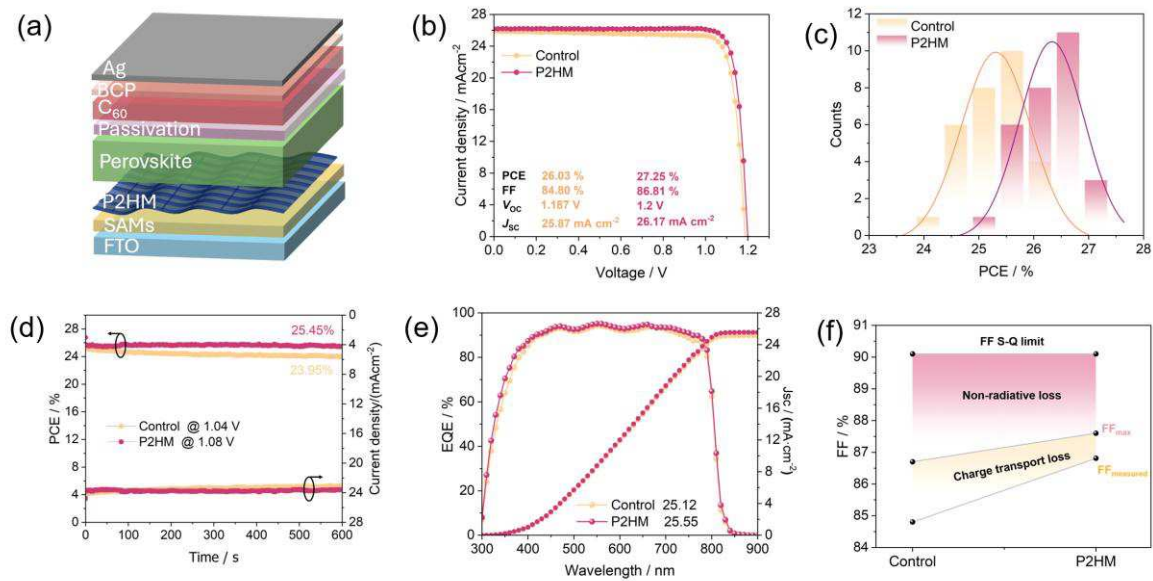
The impact of the adaptive buried interface on charge-transfer dynamics was subsequently investigated by transient absorption spectroscopy (TAS). Following excitation at 475 nm, both samples exhibit a characteristic ground-state bleaching (GSB) feature centered at approximately 776 nm (Figure 3d, e and Supplementary Figure 19). Notably, the P2HM-treated film showed a slower GSB recovery than the control film. Tracking the decay of the GSB at 776 nm showed the kinetics of the differential optical density (ΔOD) probed at the photobleaching band center (Supplementary Figure 20). The decrease in the fast component (τ_1) from 781.08 ps to 410.32 ps indicates that the interfacial hole extraction from the perovskite is enhanced. (Table S1)⁶⁰ The accelerated charge transfer is accompanied by reduced interfacial recombination, as further evidenced by enhanced steady-state photoluminescence (PL) and prolonged time-resolved photoluminescence (TRPL) lifetimes (Supplementary Figure 21 and Table S2).^{26,61}

Device-level carrier dynamics were further evaluated by transient photocurrent (TPC) and transient photovoltage (TPV) measurements (Figure 3f). The results showed that the fitted TPC lifetime decreased from 0.79 μ s (control) to 0.49 μ s (treated), indicating enhanced interfacial extraction capability. In contrast, the fitted TPV lifetime increased significantly from 27.6 μ s (control) to 52.5 μ s (treated).⁶² Collectively, these results demonstrate that the thermally activated P2HM buried interface simultaneously optimizes interfacial energetics and carrier dynamics, enabling faster hole extraction while effectively minimizing recombination losses.

Device photoelectric performance

We next evaluated the photovoltaic performance of PSCs in a p-i-n configuration incorporating a molecular confinement interface, with the device structure FTO/4PABCz/P2HM/Perovskite/Passivation layer/C₆₀/BCP/Ag (Figure 4a). Under optimized concentrations of P2HM (0.2

273 mg/mL, Supplementary Figure 22), the device achieved the champion PCE of 27.25% with a
 274 high fill factor (FF) of 86.81% and an open circuit voltage (V_{OC}) of 1.2 V (Figure 4b),
 275 substantially outperforming the control device (26.03%, FF = 84.8%, V_{OC} = 1.187 V). The
 276 treated device also exhibited less hysteresis (Supplementary Figure 23). Importantly, the
 277 P2HM-treated device achieved an independent certification PCE of 27.19% (Supplementary
 278 Figure 24).



279

280 **Figure 4** (a) Device architecture of P2HM-contained PSC. (b) $J-V$ curves of the best-
 281 performing devices measured under reverse scan direction. (c) PCE statistical distribution of
 282 Control and treated devices. (d) The corresponding SPO efficiency under their maximum
 283 power point voltage, after tracking for 10 min. (e) EQE spectra of the champion control and
 284 treated devices. (f) FF loss analysis.

285 Statistical analysis further confirms consistent improvements in FF, V_{OC} and short-circuit
 286 current density (J_{sc}) across multiple devices, demonstrating excellent reproducibility (Figure
 287 4c and Supplementary Figure 25). As shown in Figure 4d, the device treated with P2HM
 288 maintains a stable power output (SPO) of 25.45% at a bias of 1.08 V over 600 s, compared
 289 with 23.95% for the control device. The integrated J_{sc} value from the external quantum
 290 efficiency (EQE) spectrum for the treated device is 25.55 mA cm⁻² (Figure 4e), consistent with
 291 the $J-V$ characteristics. Furthermore, we verified the applicability of the P2HM layer in flexible
 292 devices. The PCE increased from 23.81% (control) to 25.05% (P2HM-treated), accompanied
 293 by a significant improvement in FF from 82.81% to 84.38% and V_{OC} from 1.184 V to 1.19 V
 294 (Supplementary Figure 26). Moreover, a high efficiency of 21.82% was achieved in flexible

modules with an area of 6.02 cm² (Supplementary Figure 27), demonstrating the versatility of P2HM.

To elucidate the origin of FF enhancement, the light intensity dependence of V_{OC} was examined. The ideality factor (n) was derived using the equation reported.⁶³ As shown in Supplementary Figure 28, compared to control ($1.39 k_B T/q$), the P2HM-treated device exhibits a lower ideality factor of $1.29 k_B T/q$, which is close to 1, indicating a more effective suppression of trap-assisted recombination. We quantified the FF improvement relative to the Shockley–Queisser (S-Q) limit by analysing losses from both non-radiative recombination and charge transport (Figure 4f). the maximum theoretical fill factor (FF_{max}) can be calculated using the Equations (2).⁶⁴

$$FF_{max} = \frac{v_{OC} - \ln(v_{OC} + 0.72)}{v_{OC} + 1} \quad (2)$$

where $v_{OC} = qV_{OC}/nk_B T$. The FF_{max} of the P2HM-treated device reaches 87.6%, which is better than that of the control device (86.7%), confirming that P2HM modification can effectively suppress nonradiative recombination and improve charge extraction efficiency.

This interpretation is further supported by electrochemical impedance spectroscopy (EIS), which shows that the recombination resistance (R_{rec}) of the P2HM-treated device increases to 35334 Ω , nearly an order of magnitude higher than that of the control device (9225 Ω) (Supplementary Figure 30, Table S3), demonstrating strongly suppressed carrier recombination.⁶⁵ Additionally, the dark $J-V$ curves (Supplementary Figure 31) further supported these findings. The trap density (N_{trap}) values were quantified using the space-charge-limited current (SCLC) method using hole-only devices (FTO/4PABCh/P2HM/Perovskite/PTAA/Ag). The values were calculated by the equation reported.⁶⁶ As shown in Supplementary Figure 32, the P2HM-treated device exhibits a markedly diminished electron N_{trap} of $6.53 \times 10^{15} \text{ cm}^{-3}$, contrasted with $9.51 \times 10^{15} \text{ cm}^{-3}$ for the control device, confirming the remarkable role of P2HM in effectively filling deep trap states.⁶⁷

Space Compatible Stability Improvement by P2HM

Finally, we investigated whether the thermally adaptive buried interface could withstand the extreme environmental stresses relevant to space photovoltaic applications. Following the ISOS stability protocols, more aggressive testing conditions were adopted to simulate the harsh thermal and radiation environments encountered in LEO. To eliminate degradation associated

with electrode corrosion, the BCP and Ag layers were replaced by a 20 nm SnO_x layer and 100 nm Au layer deposited by atomic layer deposition (ALD) and evaporator, which provides improved chemical and thermal stability.⁶⁸ When using ALD, the devices with P2HM also can achieved a PCE of 26.41% (Figure S32).

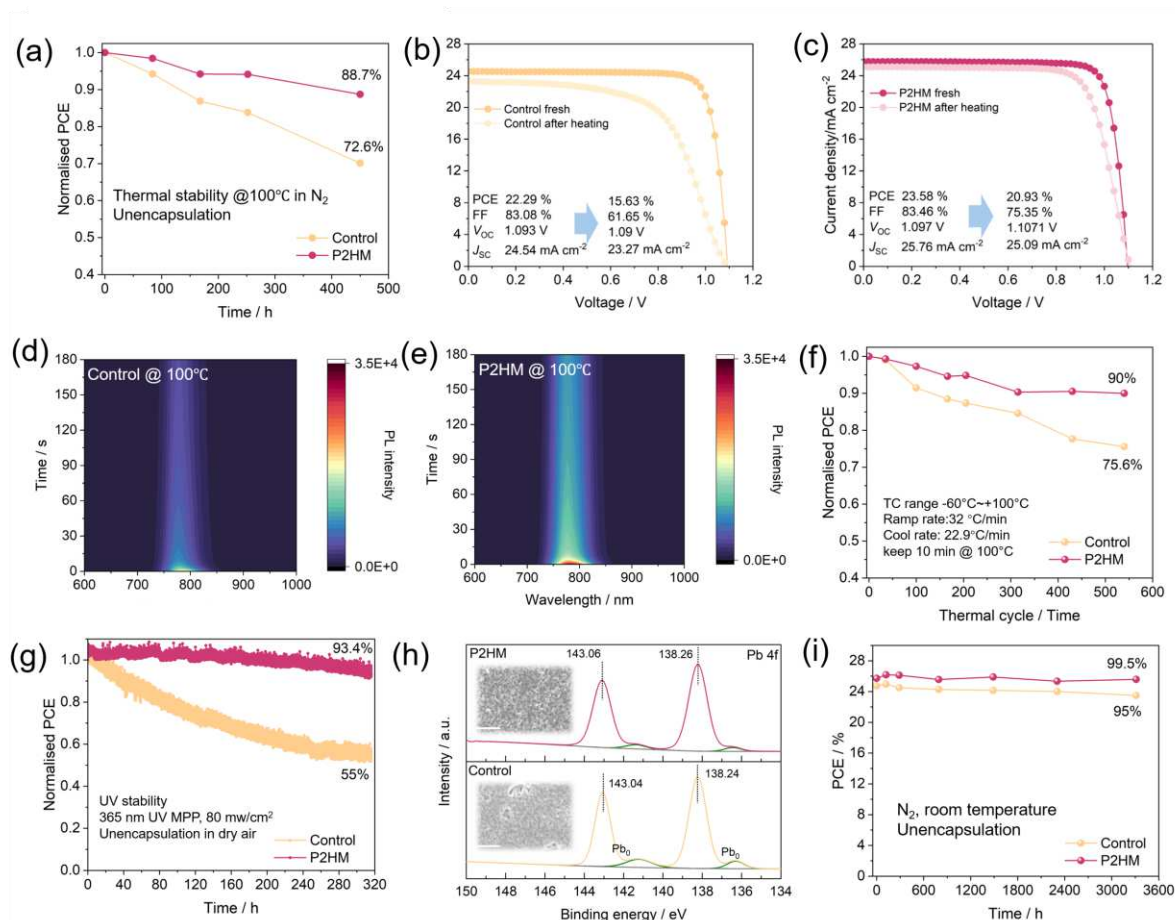


Figure 5 (a) Thermal stability of Control and P2HM-treated perovskite films. The PCE change of control (b) and treated film (c) after thermal aging. (d, e) *In-situ* PL spectra of Control and P2HM-treated films tracking at 100°C. (f) TC durability (-60°C - +100°C) of encapsulated modulus. (g) MPPT of UV exposure stability of unencapsulated devices in dry air. (h) XPS spectra and SEM images of control and treated films (bottom side) after UV aging. (i) Storage stability of unencapsulated devices under N₂.

We first evaluated the high-temperature durability of unencapsulated devices at 100 °C under nitrogen, exceeding the temperature requirement of the ISOS-D-2 protocol (85 °C). After 450 h of continuous thermal aging, the P2HM-treated device retained 88.7% of its initial PCE, substantially higher than the control device (72.6%) (Figure 5a). This is because new defects are formed at the perovskite interface during continuous high-temperature aging, leading to interface damage.³¹ Analysis of the photovoltaic parameters reveals that the degradation is

primarily associated with FF loss, whereas changes in V_{OC} and J_{SC} are comparatively limited (Figure 5b, c and Supplementary Figure 33). This behavior indicates that thermal degradation mainly originates from interface deterioration and increased defect-assisted recombination rather than bulk optical degradation. Interestingly, the V_{OC} of the P2HM-treated device increased slightly after aging, suggesting that the thermally activated polymer layer maintains effective coordination with interfacial defects during prolonged heating.⁵²

To directly examine the evolution of the perovskite film during thermal aging, quasi-Fermi level splitting (QFLS) measurements were performed. Before aging, the P2HM-treated film exhibits a higher QFLS of 1.218 V compared with 1.198 V for the control film. After 400 h at 100 °C, the QFLS values decrease to 1.060 and 1.040 V, respectively, corresponding to reductions of only 13.0% and 14.9% (Supplementary Figure 34). Moreover, buried-interface SEM images reveal severe morphological deterioration and loss of structural integrity in the control film after thermal aging, whereas the P2HM-treated interface remains substantially preserved (Supplementary Figure 35). In addition, we monitored the *in-situ* PL spectra of perovskite films during thermal aging on a hotplate at 100°C and 85°C, respectively. The P2HM-treated films exhibit a slower rate of decay (Figure 5d, e and Supplementary Figure 36, 37). These results demonstrate that the adaptive polymer layer suppresses thermally induced defect generation and structural degradation at the buried interface.

The thermal fatigue resistance was subsequently evaluated under rapid temperature cycling designed to mimic the severe thermal fluctuations experienced by low-Earth-orbit satellites. Encapsulated devices were subjected to repeated cycles between -60 °C and +100 °C, with heating and cooling rates controlled at +32 and -22.9 °C min⁻¹, respectively. They were then kept at 100°C for 10 min and at -60°C for 15 min. This accelerated protocol is substantially more demanding than the ISOS-T-3 standard (-40 to 85 °C) and better reflects space-relevant thermal shock conditions. After 540 cycles, the P2HM-treated device retained 90% of its initial efficiency, whereas the control device degraded to 75.6% (Figure 5f and Supplementary Figure 38). The dominant degradation again originates from FF deterioration, with only a 7.7% reduction for the P2HM device compared with 19.3% for the control device (Supplementary Figure 39). This enhanced thermal fatigue resistance is attributed to the improved mechanical coupling provided by the viscoelastic P2HM layer, which accommodates repeated thermal expansion and contraction while maintaining interfacial adhesion. Consistently, adhesion measurements after thermal aging reveal increased adhesion for the P2HM-treated interface, whereas the control interface suffers from adhesion loss (Supplementary Figure 40).⁶⁹ The

abundant -OH and C=O groups within P2HM establish strong intermolecular interactions with the perovskite surface, reinforcing the mechanically vulnerable buried interface.⁷⁰

Beyond thermal stresses, the influence of ultraviolet radiation was evaluated under accelerated illumination conditions. Direct photolysis of the Pb-I bond is one of the main factors triggering perovskite degradation.⁷¹ Under continuous maximum-power-point (MPP) tracking with a 365 nm UV source (85 mW cm⁻²) for 13 days, the P2HM-treated device maintains 93.4% of its initial efficiency after 13 days, while the control device decreases to 55% (Figure 5g). Furthermore, XPS testing was used to compare the chemical changes at the buried interface before and after UV testing. Compared to the results in Figure 2b, the control film shows a larger energy shift and a greater increase in Pb₀, indicating a more drastic chemical change in Pb element (Figures 5 (h) and Table S3). SEM images also show numerous voids at the buried interface of the control device after aging. These results demonstrate that the P2HM interlayer simultaneously stabilizes the chemical environment and reinforces the mechanical integrity of the buried interface, effectively mitigating multiple degradation pathways under space-relevant conditions.⁷²

The storage stability of unencapsulated devices (FTO/4PABCz/with or without P2HM/Perovskite/Passivation layer/C₆₀/BCP/Ag) were recorded in the glove box (Figure 5i). After 3300 h, the device based on P2HM layer retained 99.5% of its original PCE, while the devices based on control retained 95% of their original PCE. We also evaluated the long-term operational stability by conducting continuous operational stress at the maximum power point (MPP) under one-sun illumination. The P2HM-treated device retains 89% of its initial efficiency after 900 h, whereas the control device experiences a drop to below 48% of its initial efficiency within 220 h (Supplementary Figure 41).

Conclusion

In summary, we develop a thermally adaptive buried interface by incorporating a glass-transition-regulated P2HM layer between the SAMs and perovskite. The thermally activated molecular reconstruction of P2HM enables dynamic coordination with undercoordinated Pb²⁺, suppresses stress-induced defect formation, and simultaneously improves interfacial adhesion and charge extraction. This adaptive interface strategy delivers perovskite solar cells with a high efficiency of 27.25% and exceptional durability under space-relevant conditions, maintaining 90% of the initial efficiency after 540 rapid thermal cycles, 88.7% after prolonged

heating at 100 °C, and 93.4% after accelerated UV irradiation. These findings establish thermoresponsive buried interfaces as an effective approach for overcoming the limitations of static interface engineering and advancing efficient and reliable perovskite photovoltaics for extreme terrestrial and space applications.

Methods

Materials

All materials were obtained from commercial suppliers and used without any purification. FTO (A11-T02) was purchased from Shang Yang Technology Corp. Poly(2-hydroxyethyl methacrylate) (P2HM) was purchased from Macklin. Caesium iodide (CsI), dimethyl sulfoxide (DMSO), dimethylformamide (DMF), chlorobenzene (CB), ethyl alcohol (EtOH) and isopropyl alcohol (IPA) were purchased from Sigma Aldrich. Methylammonium chloride, methylammonium iodide (MAI) and formamidinium iodide (FAI) were purchased from Greatcell company. 1,3-Propyldiammonium diiodide (PDADI) and 3-(methylthio)propylamine hydroiodide (3MTPAI) were purchased from Xi'an Yuri Solar Co., Ltd. (4-(9'-Phenyl-9H,9'H-[3,3'-bicarbazol]-9-yl)butyl)phosphonic acid (4PABCz) was obtained from ZhouShan HuaZhou Chemical Co., Ltd. C₆₀ and bathocuproine (BCP) were purchased from Xi'an polymer light company. Lead iodide (PbI₂), was obtained from Tokyo Chemical Industry Co., Ltd. (JP). All chemicals used in this work were commercially available and were used as received.

Preparation of precursors

Stoichiometric composition of 1.5 M Cs_{0.05}FA_{0.85}MA_{0.1}PbI₃ precursor solution was acquired by dissolving 216.7 mg of FAI, 414.9 mg PbI₂ (with 10% exceed amount) and 85.8 mg MAI and MAI (10 mol%) were dissolved in 1000 µL of a mixed solution of DMF and DMSO (DMF:DMSO = 4:1). For P2HM mixture solution, different weight (0.1-0.5 mg/mL) of P2HM were added into EtOH. The SAM solution was made by dissolving 0.5 mg 4PABCz in 1 mL EtOH. The post-treatment solution was prepared by dissolving 2.5 mg 3MTPAI and 1.8 mg PDADI in 1 mL IPA/CB (1:1) mix solvents. All solutions were stirred at room temperature for 12 h and filtered through 0.22 µm PTFE filters before use.

437

438 **Fabrication of PSCs**

439 The pre-patterned FTO substrates were cleaned by sonication with detergent, deionized water,
440 acetone, and ethanol for 15 min sequentially, followed by drying with N₂ flow. Then the
441 substrates were treated with O₂-plasma for 5 min and transferred to N₂-filled glovebox
442 immediately. The 4PABCz was spin-coated at 4000 rpm for 30 s and then annealed at 100 °C
443 for 10 min. The P2HM solutions were spin-coated onto the 4PABCz dynamically at a spin
444 speed of 4000 rpm for 20 s and annealed at 100 °C for 5 min. The filtered
445 Cs_{0.05}FA_{0.85}MA_{0.1}PbI₃ precursors were deposited on the substrates by a two-step spin-coating
446 process, 1000 rpm for 10 s and then 5000 rpm for 40 s. At the last 8 s of spin-coating step, 200
447 μL of CB was dropped. Then the as-prepared perovskite films were immediately annealed at
448 100 °C for 30 min. For passivation, 60 μL of PDADI/3MPTAI solution was dynamically spin-
449 coated onto the perovskite films at 4000 rpm for 25 s and then annealed at 100°C for 5 min.
450 Finally, 25 nm C₆₀, 6 nm BCP and 100 nm Ag were sequentially deposited on the perovskite
451 films by thermal evaporation in a 1.0×10⁻⁶ Pa. vacuum chamber integrated inside the glovebox.
452 The device has an active area of 0.1 cm². For thermal related and UV stability tests, BCP was
453 replaced by SnOx (20 nm) deposited via atomic layer deposition.

454

455 **Fabrication of PSCs modules**

456 This module shares a similar structural design with the aforementioned solar cells, with the key
457 difference lying in the fabrication procedure: laser scribing is implemented to construct a series
458 configuration comprising five subcells. For the P1 isolation patterning step, Glass/ITO
459 substrates were processed via a 1064 nm laser to form P1 scribing lines with a width of 25 μm.
460 Before use, the substrates were ultrasonically cleaned sequentially in detergent solution,
461 deionized water, and ethanol, with each cleaning step lasting 15 minutes. The P2 scribing
462 process was performed subsequent to the atomic layer deposition (ALD) of tin oxide. A 532
463 nm laser was employed for P2 patterning to strip the stacked HTL/perovskite/ETL layers and
464 expose the underlying ITO electrode, yielding P2 lines with a width of 60 μm. Afterwards, a
465 100 nm-thick copper film was deposited by thermal evaporation, followed by P3 laser scribing
466 (532 nm laser) to form patterned lines with a width of 45 μm. The overall dead zone width was
467 measured to be 180 μm, corresponding to a geometric fill factor (GFF) of 95.8%.

In the fabrication of flexible solar modules, the preparation methods for all functional layers are identical to those used for rigid solar modules. A PET/ITO substrate measuring 50×50 mm² with a sheet resistance of 20 Ω is bonded to glass of the same area using OCA adhesive. Afterwards, the laminated PET/ITO is patterned with an infrared nanosecond laser operating at a repetition frequency of 600 kHz and laser power of 8% and the P1 scribing width is 25 μm. Subsequently, a green picosecond laser with 7.6% power and 600 kHz repetition frequency is adopted for P2 scribing of all layers including the HTL/perovskite/ETL stack, with a scribing width of 80 μm. Finally, following sputtering of the Cu electrode, the top electrode is patterned via a green picosecond laser set to 10% power and a repetition frequency of 500 kHz, with a P3 scribing width of 30 μm. The spacing between P1 and P2, as well as between P2 and P3, is 20 μm, and the geometric fill factor of the module was 95.9% .

Characterizations

The *J-V* curves of the PSCs were measured using a Keithley 2602B sourcemeter in an N₂-filled glovebox at room temperature under AM 1.5G condition at an intensity of 100 mW cm⁻². The calibration was performed using a standard Si solar cell (PVM937, Newport). The light source is a 450-watt xenon lamp (Oriel solar simulator, 94023A). The active area of the PSCs is 0.1 cm², defined by a metal aperture. External Quantum Efficiency (EQE) measurement was carried out by the Enlitech QE-R system under near-dark conditions. Differential scanning calorimetry (DSC) was conducted by DSC TA 250 instrument. The top-view and cross-sectional scanning electron microscope (SEM) images were scanned using a ultrahigh-resolution scanning electron microscope (Hitachi SU8600) under an accelerating voltage of 5 kV. The X-ray diffraction (XRD) spectra were recorded by an X-ray diffract meter (Bruker D8 Advance) with a monochromatic Cu-Kα X-ray source. Grazing-incidence X-ray diffraction (GIXRD) measurements were performed using an X'Pert Powder diffractometer (PANalytical, The Netherlands) equipped with a Cu Kα radiation source ($K\alpha_1 = 1.540598 \text{ \AA}$, $K\alpha_2 = 1.544426 \text{ \AA}$, $K\alpha = 1.541874 \text{ \AA}$), operated at 40 kV and 40 mA. The Grazing-incidence wide-angle X-ray scattering (GIWAXS) was performed at the Shanghai Synchrotron Radiation Facility (SSRF) using an X-ray with a wavelength of 1.24 Å. Ultrafast transient absorption (TA) spectroscopy was carried out using a HARPIA-TA system (Light Conversion, Lithuania) operating at a repetition rate of 33 kHz with an average laser output power of 10 W. Steady state photoluminescence (PL) spectra were measured in ambient air at room temperature by a

photoluminescence spectrometer (Edinburgh FLS1000), with the samples illuminated from the film side. In-situ PL measurements were carried out using an InView-PL device from Shanghai Fuxiang Optical Co. Ltd. and the model of the spectrometer was PG2000-Pro. X-ray photoelectron spectroscopy (XPS) measurements were carried out under ultra-high-vacuum (UHV) conditions ($<10^{-8}$ mbar) using a monochromatic Al K α source ($h\nu = 1486.6$ eV) on an ESCA 2SR high-throughput spectrometer. Ultraviolet photoelectron spectroscopy (UPS) measurements were performed on a Nexsa spectrometer (Thermo Fisher Scientific, USA) using a He I ultraviolet source (21.22 eV) with a spot size of 3×3 mm² under an ultrahigh vacuum ($<2 \times 10^{-9}$ mbar). The valence band spectra and work function were measured without and with a sample bias of -5 or -10 V, respectively. Atomic force microscope (AFM) morphology and kelvin probe force microscopy (KPFM) measurements were conducted on the FastScan Bio (Bruker). Maximum power point (MPP) tracking was tested using white LED at 100 mW cm⁻² without UV filter. Samples were under illumination in an N₂ or dry air environment at 25 ± 5 °C without any encapsulation.

DFT Calculations

All electrostatic potential (ESP) computations were performed using the Gaussian 16 (revision C.01) suite. Throughout the study, the PBE0 hybrid exchange–correlation functional was employed, together with Grimme’s D3 dispersion correction with Becke–Johnson damping (DFT-D3BJ).⁷³ Geometry optimizations and vibrational analyses were conducted with the def2-SVP basis set.^{74,75} Single-point electronic energies were subsequently refined using the def2-TZVP basis set. ESP surfaces were visualized with the help of the Multiwfn and VMD software.⁷⁶

The first-principles were employed to carry out the spin-polarization DFT calculations and all spin polarized DFT calculations were performed with the Perdew-Burke-Ernzerhof PBE exchange-correlation functional in a plane wave pseudopotential implementation using the Vienna ab initio simulation packages (VASP)^{77,78}. The projector augmented wave (PAW) method was employed to describe the core–valence interaction,⁷⁹ with a plane-wave cutoff energy of 450 eV. Electron smearing was employed through the Gaussian smearing method with a smearing width of 0.05 eV. The $1 \times 1 \times 1$ k -point mesh was used in all calculations to sample the Brillouin zone integration. The van der Waals force was considered in these calculations via the DFT-D3 correction method. The convergence criterion for the electronic

self-consistent iteration was set to 10^{-5} eV, and the maximum force on each atom was constrained to be less than 0.03 eV/Å. The spin polarization was considered in all calculation.

Acknowledgements

The authors acknowledge funding support from the National Natural Science Foundation of China (No.62274040 and No.62304046), Shanghai science and technology innovation action plan (No. 24DZ3001200) and Shanghai Science and Technology Innovation Action Plan 2023 Special Project for Supporting Carbon Peak Carbon Neutrality Project (23DZ1200400).

We thank the staff members at BL19U2 beamline of the National Facility for Protein Science in Shanghai (NFPS), Shanghai Advanced Research Institute, Chinese Academy of Sciences for providing technical support and assistance in data collection and analysis.

Conflicts of interest

There are no conflicts to declare

Contributions

R.W. conceived the idea, designed the experiments, performed the device fabrication and characterization, analysed the data, and wrote the manuscript. L.D. and X.L. fabricated the perovskite modules and helped with stability tests. Z.H. contributed to the optimization of the perovskite solar cells. H.Z. performed the C-AFM measurements. D.Z. carried out the XPS characterization. D.L. performed the TA measurements. L.X. conducted the SS ^1H NMR measurements. Q.G. helped with the *in-situ* PL measurements. T.L. assisted with the thermal evaporation process. S.H. offered help with XRD characterization. Y.G. offered help in GIWAXS characterization. J.W. helped with crystallization analysis. A.Y. provided valuable suggestions for the manuscript. Y.Z. supervised the project. All authors discussed the results and contributed to the revision of the manuscript.

- 560 1 Miao, T. *et al.* An entropy-regulating molecular lock stabilizes formamidinium lead
561 halide perovskite. *Science* **392**, 742-748 (2026).
- 562 2 Wu, X. *et al.* Electronic-resonance enhanced molecule for perovskite solar cells. *Nature*,
563 1-3 (2026).
- 564 3 Li, T. *et al.* Quasi-random oriented molecular contacts for inverted perovskite solar cells
565 with improved efficiency. *Nat. Energy*, 1-10 (2026).
- 566 4 Jinno, H. *et al.* Ultrathin Perovskite Solar Cells With γ -Ray Tolerance Enabled by a
567 Flexible Radiation-Resistant Plastic Substrate. *Sol. RRL* **10**, e70339 (2026).
- 568 5 Guo, J. *et al.* AI-guided design of efficient perovskite solar cells operationally stable at
569 100° C. *Science* **392**, 724-728 (2026).
- 570 6 Lee, M. *et al.* Towards space compatible perovskite solar cells: guidelines for thermal
571 shock resilience and near space balloon testing. *Energy Environ. Sci.* **19**, 2557-2568
572 (2026).
- 573 7 Testing, A. S. f., Simulation, M. C. E. o. S. & Technology, A. o. S. *Standard solar*
574 *constant and zero air mass solar spectral irradiance tables*. (ASTM International,
575 2000).
- 576 8 Bush, M. E. *et al.* Space environment considerations for perovskite solar cell operations:
577 A review. *Acta Astronaut.* **235**, 235-250 (2025).
- 578 9 Yun, D. *et al.* Interfacial stability of organic-based hole transport layers in perovskite
579 photovoltaics for space-like thermal environments. *Comm. Eng.* (2026).
- 580 10 NOAA-STAR, NOAA-21 satellite data, <[https://ncc.nesdis.noaa.gov/NOAA-](https://ncc.nesdis.noaa.gov/NOAA-21/index.php)
581 [21/index.php](https://ncc.nesdis.noaa.gov/NOAA-21/index.php)> (
- 582 11 IEC 60068-2-14:2023, <<https://webstore.iec.ch/en/publication/71503>> (
- 583 12 Fei, C. *et al.* Strong-bonding hole-transport layers reduce ultraviolet degradation of
584 perovskite solar cells. *Science* **384**, 1126-1134 (2024).
- 585 13 Liu, D. *et al.* Strain analysis and engineering in halide perovskite photovoltaics. *Nat.*
586 *Mater.* **20**, 1337-1346 (2021).
- 587 14 Bamisile, O., Acen, C., Cai, D., Huang, Q. & Staffell, I. The environmental factors
588 affecting solar photovoltaic output. *Renew. Sustain. Energy Rev.* **208**, 115073 (2025).
- 589 15 Wu, L. *et al.* Resilience pathways for halide perovskite photovoltaics under temperature
590 cycling. *Nat. Rev. Mater.* **10**, 536-549 (2025).
- 591 16 Zhong, Y. *et al.* Homogenizing out-of-plane strain distribution for high-performance
592 flexible perovskite photovoltaics. *Sci. Adv.* **12**, eaec3238 (2026).
- 593 17 Erdil, U. *et al.* Delamination of perovskite solar cells in thermal cycling and outdoor
594 tests. *Energy Technol.* **13**, 2401280 (2025).
- 595 18 Xiong, Z. *et al.* Electrodeposited self-assembled molecules for perovskite photovoltaics.
596 *Nature*, 1-2 (2026).
- 597 19 Chen, M. *et al.* Stress engineering for mitigating thermal cycling fatigue in perovskite
598 photovoltaics. *ACS Energy Lett.* **9**, 2582-2589 (2024).
- 599 20 Li, Z. *et al.* Stabilized hole-selective layer for high-performance inverted pin perovskite
600 solar cells. *Science* **382**, 284-289 (2023).
- 601 21 Liu, F. *et al.* Misplaced-dipole engineered repairable fluoropolymer elastomer for
602 flexible perovskite solar cell with excellent thermal-mechanical cycling resistance. *Nat.*
603 *Commun.* (2026).
- 604 22 Zhao, L. *et al.* Wide-temperature-range fluid-phase grain boundaries for temperature-
605 robust perovskite solar cells. *Nat. Photon.*, 1-9 (2026).

- 23 Zhang, Z. *et al.* Photoswitchable isomers to improve grain boundary resilience and perovskite solar cells stability under light cycling. *Nat. Energy* **11**, 623-632 (2026). <https://doi.org/10.1038/s41560-026-01993-z>
- 24 Shao, C. *et al.* Strain release via glass transition temperature regulation for efficient and stable perovskite solar cells. *Adv. Mater.* **37**, 2417150 (2025).
- 25 Li, L. *et al.* Buried Interface Chelating Molecular Bridge Strategy Enables Highly Efficient and Stable Inverted Perovskite Solar Cells. *Adv. Mater.* **38** (2025). <https://doi.org/10.1002/adma.202518406>
- 26 Wu, Y. *et al.* Chemically coupled dual-interface self-regulation for perovskite solar cells. *Nat. Commun.* (2026).
- 27 Song, F. *et al.* Buried interface modification for high performance and stable inverted perovskite solar cells. *Angew. Chem. Int. Ed.* **64**, e202516012 (2025).
- 28 Zhang, Y. *et al.* Improved fatigue behaviour of perovskite solar cells with an interfacial starch–polyiodide buffer layer. *Nat. Photon.* **17**, 1066-1073 (2023).
- 29 Yilmaz, C. *et al.* Perovskite solar cells with enhanced thermal fatigue resistance under extreme temperature cycling. *Nat. Commun.* **17**, 3669 (2026).
- 30 Li, G. *et al.* Highly efficient pin perovskite solar cells that endure temperature variations. *Science* **379**, 399-403 (2023).
- 31 Zhang, Y. *et al.* Thermo-Mechanically Robust Inverted Perovskite Solar Cells Toward Extreme Environments. *Adv. Funct. Mater.* **36**, e75697 (2026).
- 32 Dong, B. *et al.* Self-assembled bilayer for perovskite solar cells with improved tolerance against thermal stresses. *Nat. Energy* **10**, 342-353 (2025).
- 33 Zhang, J. *et al.* In situ dynamic regulation of strain at the buried interface of stable perovskite solar cells. *Nat. Photon.* **20**, 119-127 (2026).
- 34 Peng, Y. *et al.* Enlarging moment and regulating orientation of buried interfacial dipole for efficient inverted perovskite solar cells. *Nat. Commun.* **16**, 1252 (2025).
- 35 Ye, C. *et al.* Structural dynamics of melting and glass formation in a two-dimensional hybrid perovskite. *Nat. Commun.* **16**, 7696 (2025).
- 36 Tsiptsias, C. Thermochemical transition in non-hydrogen-bonded polymers and theory of latent decomposition. *Polymers* **14**, 5054 (2022).
- 37 Williams, M. L., Landel, R. F. & Ferry, J. D. The temperature dependence of relaxation mechanisms in amorphous polymers and other glass-forming liquids. *J. Am. Chem. Soc.* **77**, 3701-3707 (1955).
- 38 Katagiri, S., Nagata, H. & Honda, H. Solid-State NMR Studies of Hydrated and Dehydrated States of Temperature-Responsive Polymers. *ACS Appl. Polym. Mater.* **7**, 10919-10929 (2025).
- 39 Sun, R. *et al.* A stepwise melting-polymerizing molecule for hydrophobic grain-scale encapsulated perovskite solar cell. *Adv. Mater.* **37**, 2410395 (2025).
- 40 Kuo, S.-W., Lin, C.-L. & Chang, F.-C. Phase behavior and hydrogen bonding in ternary polymer blends of phenolic resin/poly (ethylene oxide)/poly (ϵ -caprolactone). *Macromolecules* **35**, 278-285 (2002).
- 41 Zhuang, R. *et al.* Co-deposited inverted perovskite photovoltaics towards 27% efficiency via vertical redistribution of self-assembled-molecules and in-situ crosslinking. *Nat. Commun.* (2026).
- 42 Zhao, Y. *et al.* Enhanced interface adhesion with a polymeric hole transporter enabling high-performance air-processed perovskite solar cells. *Energy Environ. Sci.* **18**, 1366-1374 (2025).
- 43 Ren, L. *et al.* Reversible Thermally Driven Phenyl Torsion in Self-Assembled Monolayers Releases Strain for Heat-Resilient Inverted Perovskite Solar Cells. *Angew. Chem. Int. Ed.* **65**, e21774 (2026).

656 44 Bi, D. *et al.* Polymer-templated nucleation and crystal growth of perovskite films for
657 solar cells with efficiency greater than 21%. *Nat. Energy* **1**, 1-5 (2016).

658 45 Han, T.-H. *et al.* Perovskite-polymer composite cross-linker approach for highly-stable
659 and efficient perovskite solar cells. *Nat. Commun.* **10**, 520 (2019).

660 46 Chen, L. *et al.* Ionic bonding without directionality facilitates efficient interfacial
661 bridging for perovskite solar cells. *Small* **20**, 2308964 (2024).

662 47 Wang, J. *et al.* Bimolecular Crystallization Modulation Boosts the Efficiency and
663 Stability of Methylammonium-Free Tin–Lead Perovskite and All-Perovskite Tandem
664 Solar Cells. *Adv. Energy Mater.* **14**, 2402171 (2024).

665 48 Wang, K. *et al.* Amidino-based ligand enables oriented crystallization and boosted
666 carrier extraction for inverted perovskite solar cells and modules. *Joule* **9** (2025).

667 49 Jia, Z. *et al.* Film-forming polymer nanoparticle strategy for improving the passivation
668 and stability of perovskite solar cells. *Energy Environ. Sci.* **17**, 7221-7233 (2024).

669 50 Chen, H. *et al.* Improved charge extraction in inverted perovskite solar cells with dual-
670 site-binding ligands. *Science* **384**, 189-193 (2024).

671 51 Yang, Y. *et al.* Multifunctional Conjugated Ligands Synergistically Optimize the
672 Interface and Regulate Crystallization for High-Performance 2D/3D Perovskite Solar
673 Cells. *Adv. Mater.* **38**, e20251 (2026).

674 52 Huang, S. *et al.* Molecularly Engineered Self-Healing Scaffold With Customized
675 Dynamic Bonds Enable Stable and Scalable Flexible Perovskite Solar Cells and
676 Modules. *Adv. Mater.*, e73855 (2026).

677 53 Wang, F. *et al.* Enhancing Heterogeneous Nucleation on Buried Interface for Efficient
678 Antisolvent-Free Inverted Flexible Perovskite Photovoltaics. *Adv. Mater.*, e73358
679 (2026).

680 54 Yang, G. *et al.* Molecular confinement for enhanced interfacial contact and performance
681 in inverted perovskite solar cells. *Nat. Commun.* (2026).

682 55 Li, Y. *et al.* Localized Lead-Chelating Insulator Bottom Contact for Efficient and Stable
683 p-i-n Perovskite Solar Cells. *Adv. Sci.* **12**, e09816 (2025).

684 56 Xing, C. *et al.* Exploring a Scalable Route for Efficient Flexible Perovskite Solar Cells
685 via Amphiphilic Cross-linkable Monomer. *Energy Environ. Sci.* (2026).

686 57 Xu, P. *et al.* Retarding the growth kinetics of chemical bath deposited nickel oxide films
687 for efficient inverted perovskite solar cells and minimodules. *Adv. Mater.* **37**, 2505087
688 (2025).

689 58 Sun, X. *et al.* Multifunctional Modification of the Buried Interface in Mixed Tin-Lead
690 Perovskite Solar Cells. *Angew. Chem. Int. Ed.* **63**, e202409330 (2024).

691 59 Li, T. *et al.* Inorganic wide-bandgap perovskite subcells with dipole bridge for all-
692 perovskite tandems. *Nat. Energy* **8**, 610-620 (2023).

693 60 Li, C. *et al.* Photostable wide-bandgap perovskites with enhanced interface coupling
694 for all-perovskite tandems. *Energy Environ. Sci.* (2026).

695 61 Wang, R. *et al.* Improving the efficiency and stability of perovskite solar cells using π -
696 conjugated aromatic additives with differing hydrophobicities. *Energy Environ. Sci.* **16**,
697 2646-2657 (2023).

698 62 Wang, R. *et al.* Binary Additives Synergistically Modulate Crystallization for Enhanced
699 HTL-Free Sn–Pb Perovskite Solar Cells. *Adv. Energy Mater.*, e71084 (2026).

700 63 Qu, Z. *et al.* Enhanced charge carrier transport and defects mitigation of passivation
701 layer for efficient perovskite solar cells. *Nat. Commun.* **15**, 8620 (2024).

702 64 Cao, Q. *et al.* Meticulous Construction of Internal Encapsulation Layer via In Situ Self-
703 Cross-Linking Polymerization and Ring-Opening Addition Reactions for Efficient and
704 Environmental Perovskite Solar Cells. *Adv. Mater.* **38**, e17304 (2026).

- 65 Cao, X. *et al.* Full-Cycle Humidity-Tolerant Perovskite Solar Cells Enabled by Air-
66 Processed In Situ Shielding via Synergistic Dynamic Bonds and Self-Crosslinking. *Adv.*
67 *Energy Mater.* **15**, e03437 (2025).
- 66 Wang, X. *et al.* Elimination of charge accumulation by a self-assembled cocrystal
69 interlayer for efficient and stable perovskite solar cells. *Energy Environ. Sci.* **17**, 569-
710 579 (2024).
- 67 Shen, H. *et al.* Homogenizing interfacial assembly via indole-mediated binary
712 monolayers for perovskite solar cells. *Nat. Commun.* (2026).
- 68 Wang, M. *et al.* Decoupling cation segregation and volatile loss in formamidinium-
714 caesium metal halide perovskite solar cells under high-temperature operating
715 conditions. *Nat. Energy*, 1-12 (2026).
- 69 Dai, Z. *et al.* Interfacial toughening with self-assembled monolayers enhances
717 perovskite solar cell reliability. *Science* **372**, 618-622 (2021).
- 70 Li, Z. *et al.* Hyperbranched polymer functionalized flexible perovskite solar cells with
719 mechanical robustness and reduced lead leakage. *Nat. Commun.* **14**, 6451 (2023).
- 71 Chen, X. *et al.* Photo-Tautomerization-Driven Energy Transfer at the Hole-Transport
721 Interface Stabilizes Efficient Inverted Perovskite Solar Cells. *Angew. Chem. Int. Ed.* **65**,
722 e3596437 (2026).
- 72 Tao, M. *et al.* Molecule-triggered strain regulation and interfacial passivation for
724 efficient inverted perovskite solar cells. *Joule* **8**, 3142-3152 (2024).
- 73 Grimme, S., Ehrlich, S. & Goerigk, L. Effect of the damping function in dispersion
726 corrected density functional theory. *J. Comput. Chem.* **32**, 1456-1465 (2011).
- 74 Weigend, F. & Ahlrichs, R. Balanced basis sets of split valence, triple zeta valence and
728 quadruple zeta valence quality for H to Rn: Design and assessment of accuracy. *Phys.*
729 *Chem. Chem. Phys.* **7**, 3297-3305 (2005).
- 75 Scalmani, G. & Frisch, M. J. Continuous surface charge polarizable continuum models
731 of solvation. I. General formalism. *J. Chem. Phys.* **132** (2010).
- 76 Lu, T. & Chen, F. Multiwfn: A multifunctional wavefunction analyzer. *J. Comput. Chem.*
733 **33**, 580-592 (2012).
- 77 Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made
735 simple. *Phys. Rev. Lett.* **77**, 3865 (1996).
- 78 Kresse, G. & Furthmüller, J. Efficiency of ab-initio total energy calculations for metals
737 and semiconductors using a plane-wave basis set. *Comput. Mater.* **6**, 15-50 (1996).
- 79 Kresse, G. & Joubert, D. From ultrasoft pseudopotentials to the projector augmented-
739 wave method. *Phys. Rev. B* **59**, 1758 (1999).

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [ESINPRanWang25082026.docx](#)