Intermediate Phase Modulation and Defect Passivation to Stabilize the Photoactive Phase of Formamidinium-Based Perovskites for Air-Ambient Device Fabrication

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Rationale for chain-length selection of alkyl amine additives:

The selection of n-heptylamine (C7) as the amine additive was motivated by the need to balance steric hindrance, molecular mobility, and electronic compatibility during perovskite crystallization. Short-chain alkyl amines (e.g., C4–C5) provide limited steric protection and weaker hydrophobic passivation, resulting in insufficient suppression of defect formation. In contrast, longer alkyl chains (≥C8) tend to introduce excessive insulating character, which can impede charge transport and disrupt crystallization continuity. A C7 alkyl chain offers an optimal compromise, enabling effective coordination of the terminal $–NH_2$ group with Pb^{2+} ions while maintaining favorable film morphology, crystallization kinetics, and interfacial stability, as supported by previous reports on alkyl amine and spacer-assisted perovskite growth.[1-3]

Table: S1 Circuit fitting parameters of TRPL for carrier life time calculation.

Specification	D_1 (%)	τ_1 (ns)	D_2 (%)	τ_2 (ns)	τ_{avg} (ns)
CB	72	98.5	28	670.3	259.13
n-HA + CB	38	285.7	62	1125.6	814.40

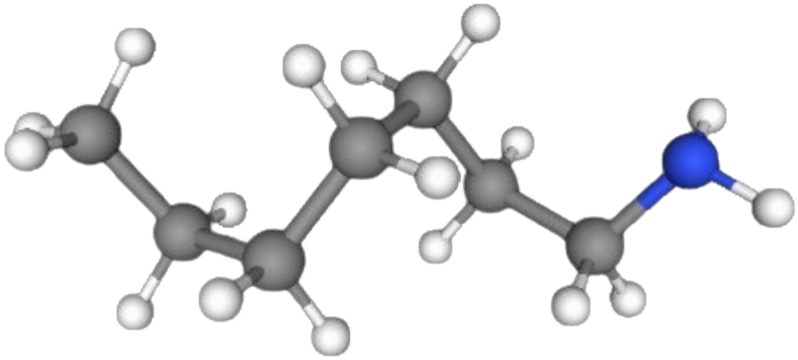

Figure S1: *Schematic illustration of the molecular structure of n-heptylamine (n-HA), highlighting the terminal amine group and the linear C7 alkyl chain*

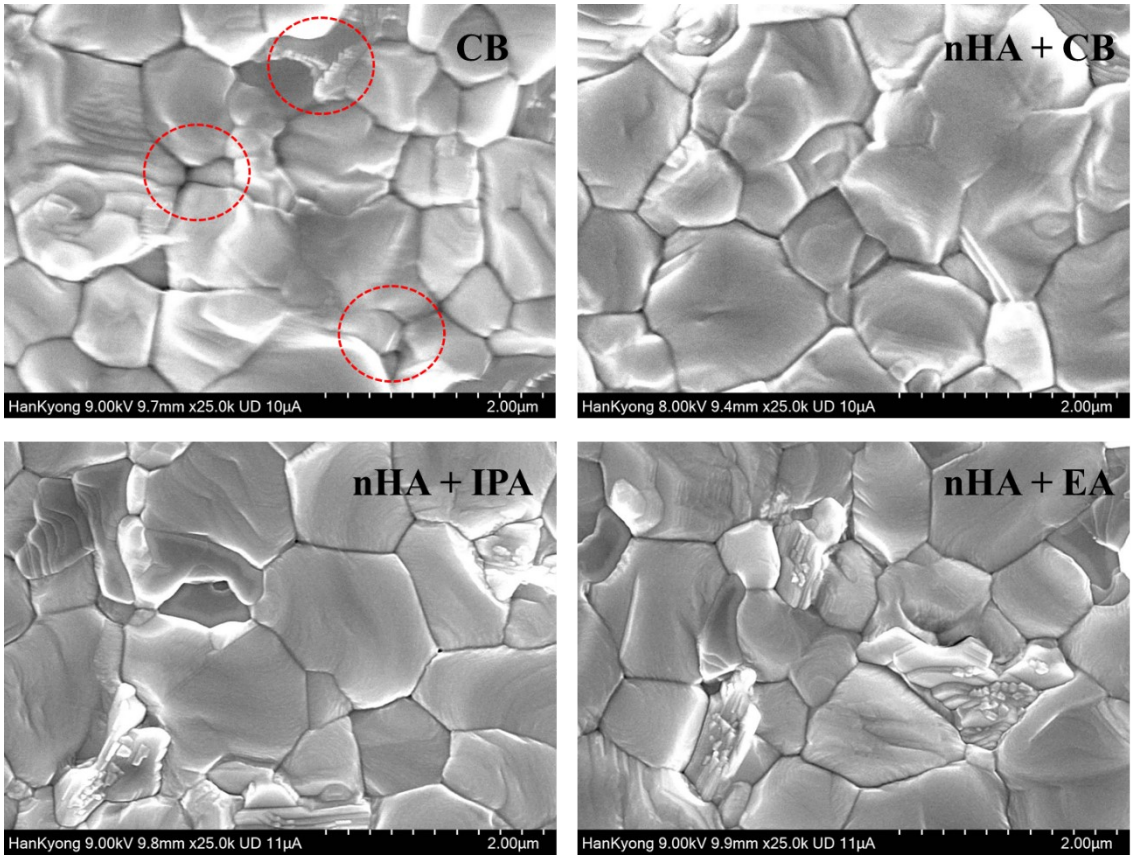

Figure S2: *Top view SEM of perovskite films with various antisolvent combinations.*

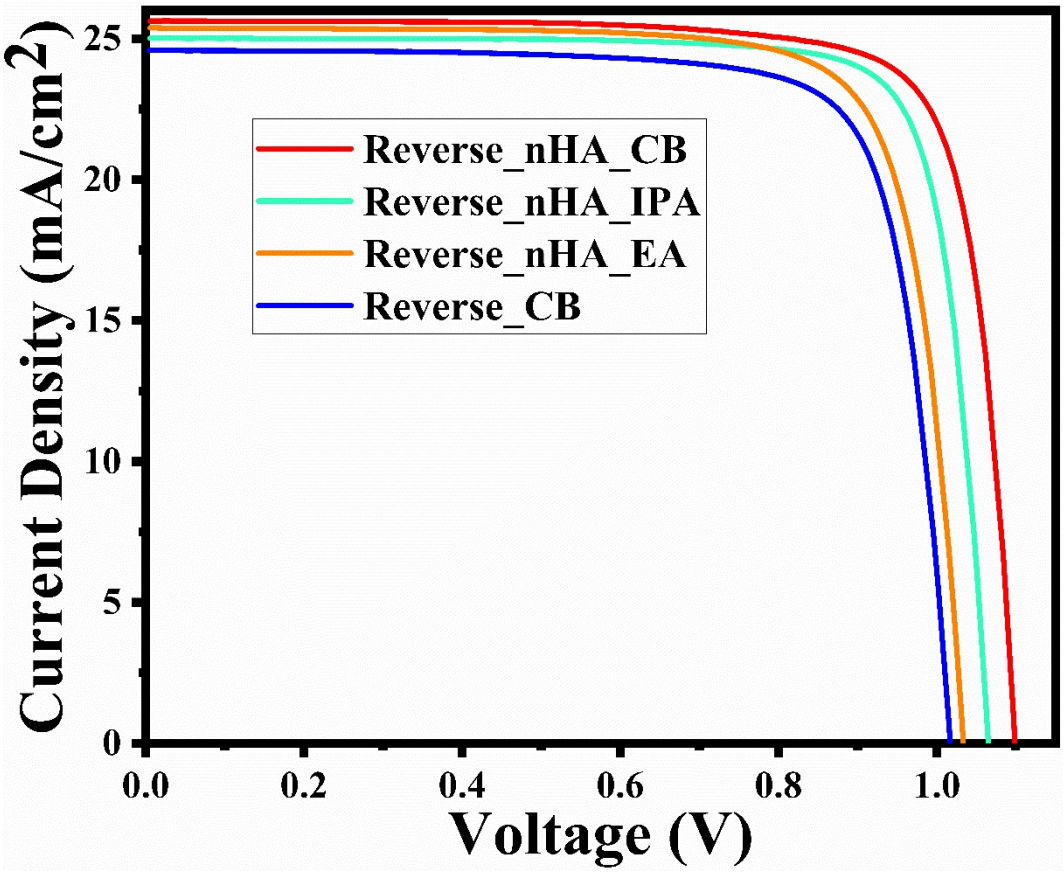

Figure S3: *Reverse scan JV curves for various antisolvent combinations.*

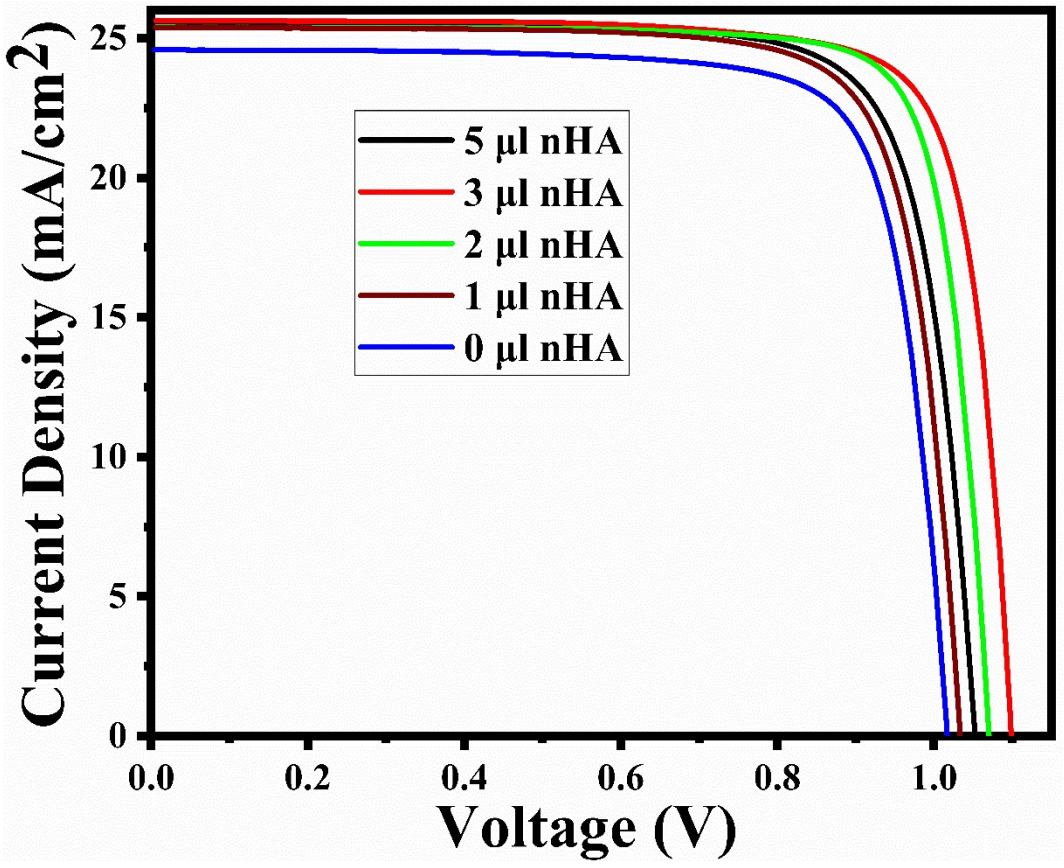

Figure S4: *Concentration optimization of n-HA mixed in CB as antisolvent.*

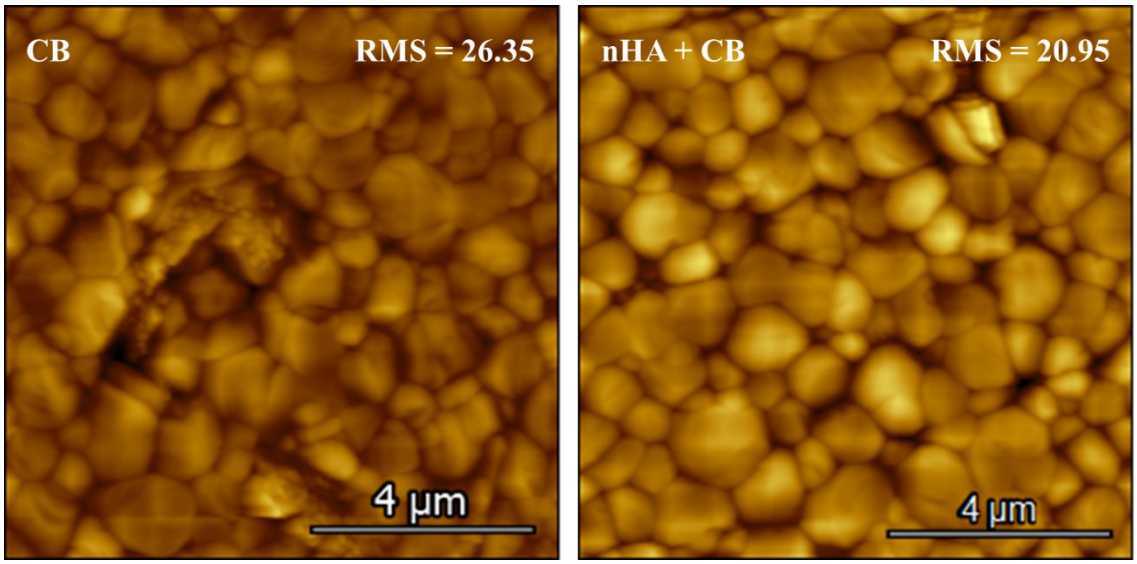

Figure S5: *AFM of pristine and modified perovskite films.*

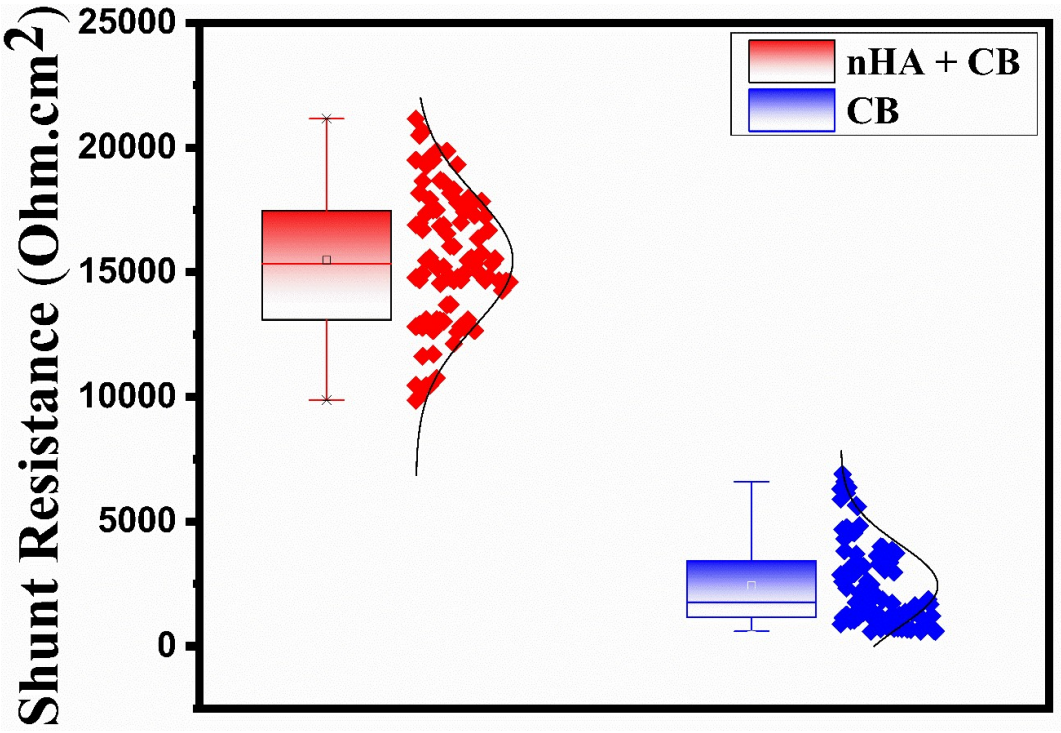

Figure S6: *Effect of modification on the distribution of shunt resistance.*

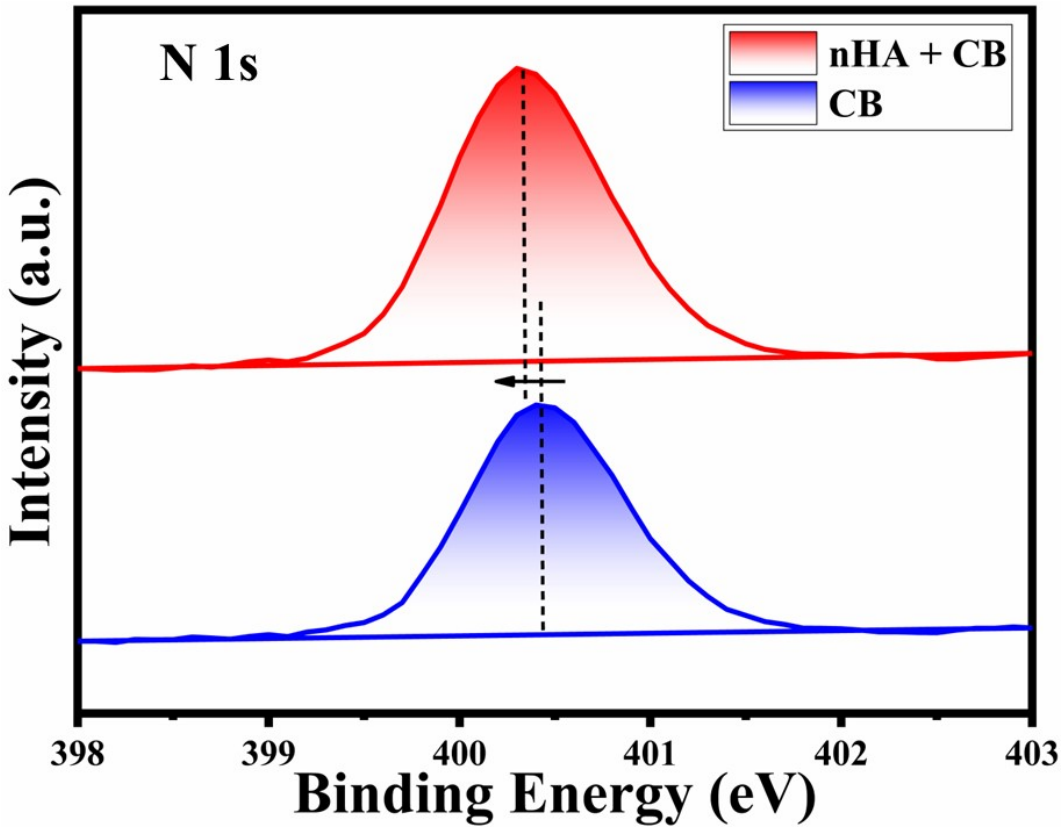

Figure S7: *N1s peak of perovskite film from XPS elemental analysis.*

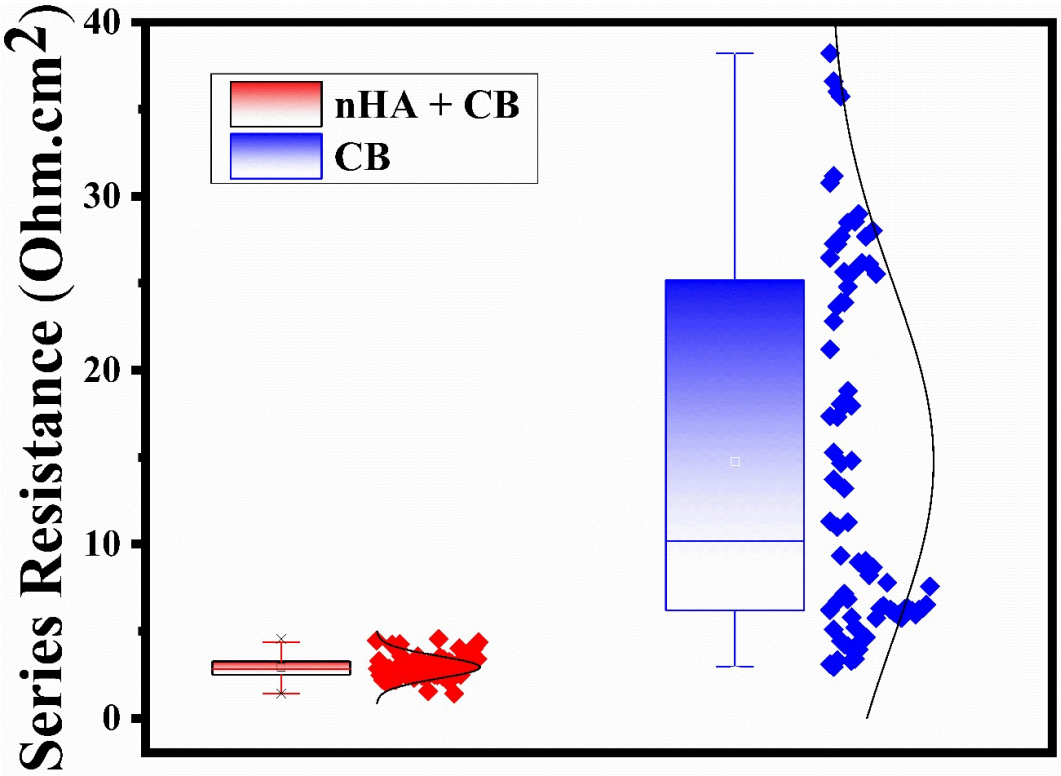

Figure S8: *Effect of modification on the distribution of series resistance of devices.*

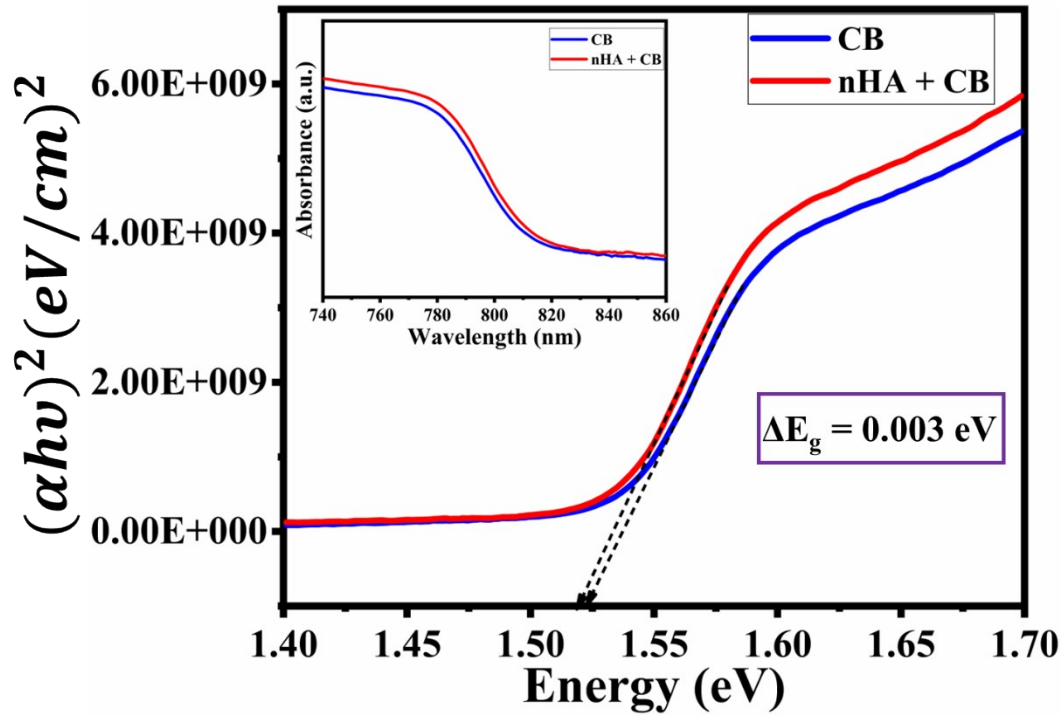

Figure S9: *Tauc plot analysis of pristine and modified perovskite films (**Inset:** UV-Vis absorbance spectra of perovskite films).*

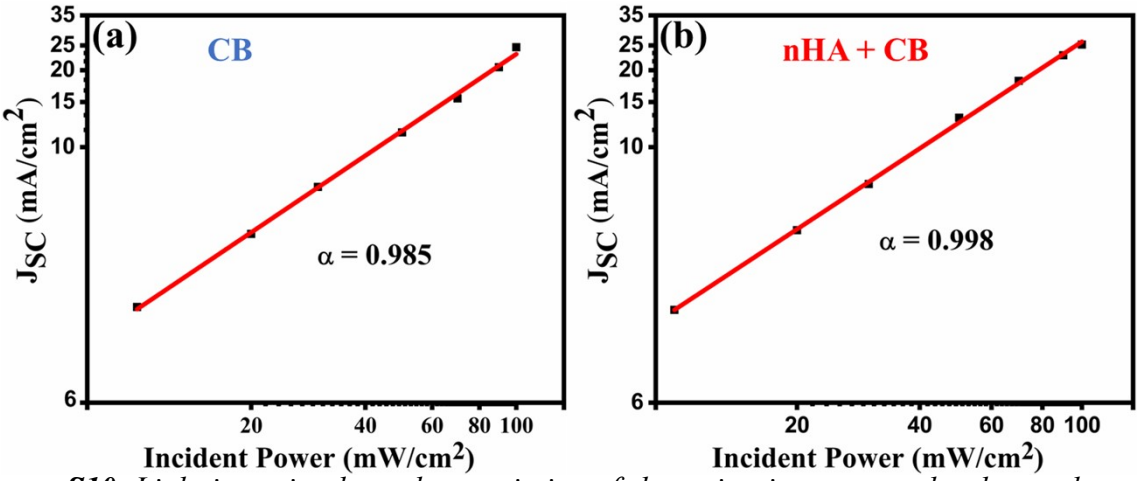

Figure S10: *Light intensity-dependent variation of short circuit current at log-log scale.*

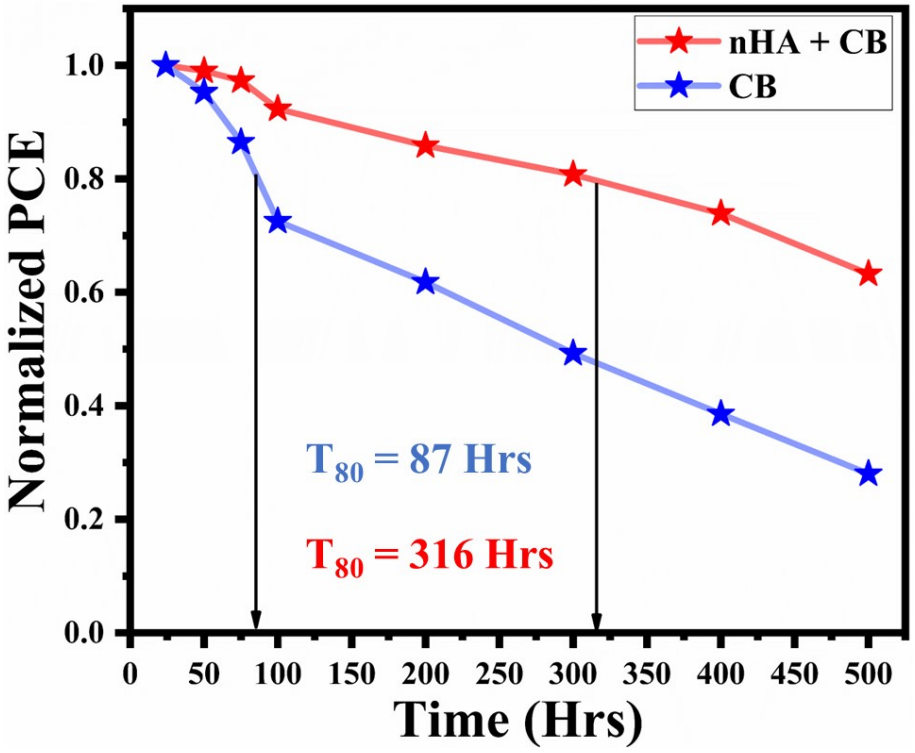

Figure S11: *Stability analysis of perovskite devices at the elevated humidity value.*

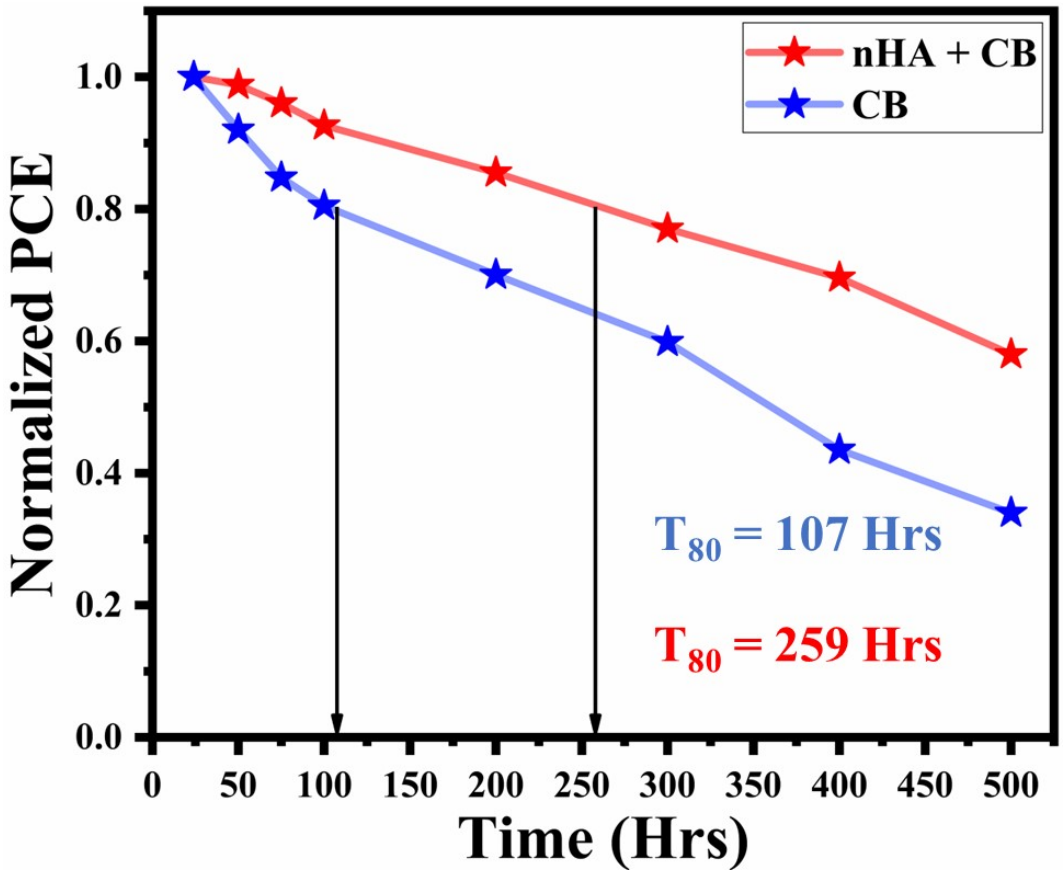

Figure S12: *Stability analysis of perovskite devices at the elevated temperature value.*

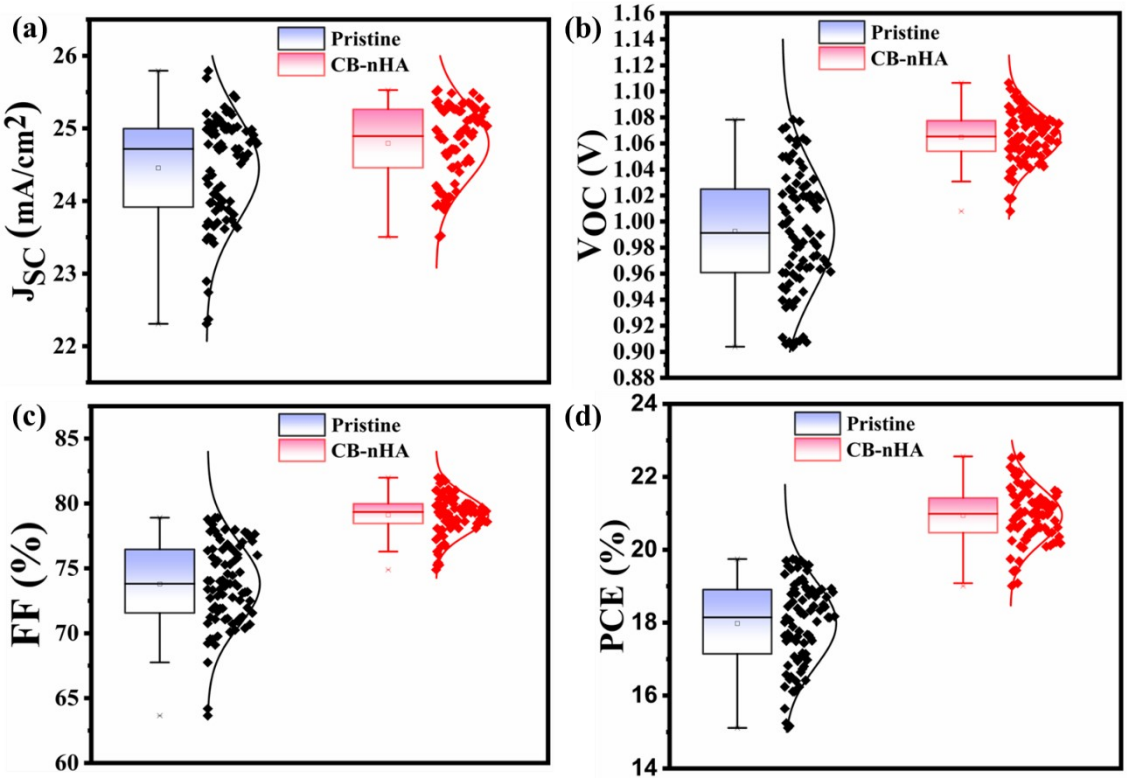

Figure S13: *Distribution of performance parameters for pristine and modified devices.*

References:

1. H. Nagasaka, M. Yoshizawa-Fujita, Y. Takeoka and M. Rikukawa, *ACS Omega*, 2018, **3**, 18925-18929.
2. Y. Tezuka, K. Umemoto, M. Takeda, Y. Takahashi, H. Ebe, J. Enomoto, S. Rodbuntum, T. Nohara, D. Fontecha, S. Asakura, T. Chiba, M. I. Furis, T. Yoshida, H. Uji-i and A. Masuhara, *Japanese Journal of Applied Physics*, 2020, **59**, SDDC04.
3. P. Zhao, J. Subbiah, B. Zhang, J. A. Hutchison, G. Ahluwalia, V. Mitchell, K. P. Ghiggino and D. J. Jones, *Advanced Materials Interfaces*, 2023, **10**, 2202313.