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Supporting Information

Boosting fill factor and open-circuit voltage of carbon-based perovskite solar cells with graphene co-doped P3HT/NiO_x hole-transporting bilayer

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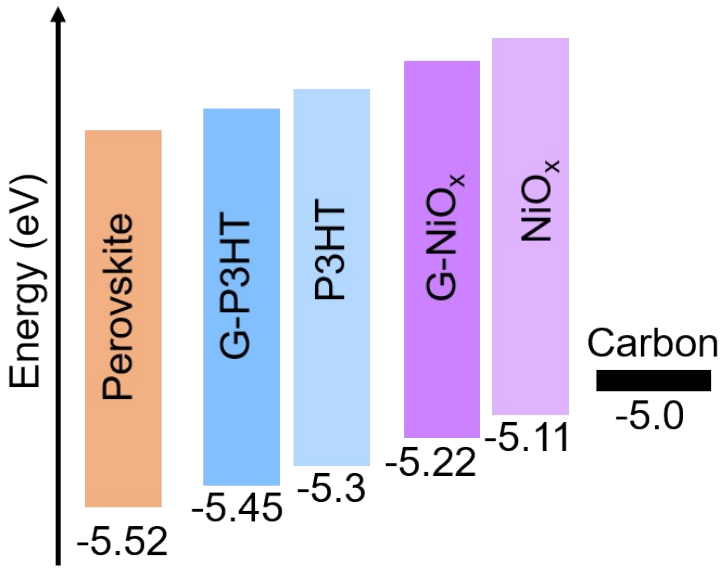

Figure S1. Energy level diagrams of perovskite, G-P3HT, P3HT, G-NiO_x, NiO_x, and carbon electrode, wherein the HOMO value of pristine P3HT and the VBM value of NiO_x are reproduced from our previous work (*Adv. Funct. Mater., 2023, 33, 2301920*).

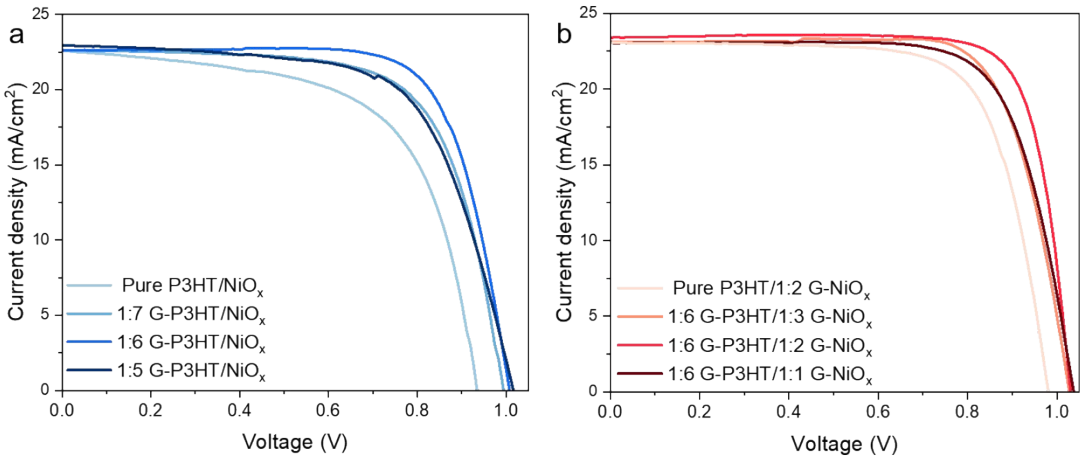

Figure S2. J-V curves of C-PSCs with (a) different graphene-doped P3HT and (b) different graphene-doped NiO_x HTLs. It can be found in Figure S2a that doping graphene into P3HT can significantly improve the device performance, especially V_{oc} and FF. The optimal volume ratio of graphene and P3HT solution is 1: 6. This improvement is mainly attributed the increased conductivity and decreased HOMO level of P3HT by graphene doping. Based on this result, we further explore the effect of graphene concentration in NiO_x on C-PSC performance, as shown in Figure S2b. It is obvious that graphene doping in NiO_x also increases the device efficiency, mainly FF, which is due to the enhanced electricity contact between carbon electrode and G-NiO_x. With the volume ratio of 1: 2 between graphene and NiO_x solution, the resultant G-P3HT/G-NiO_x devices yield the highest efficiency of 19.1%.

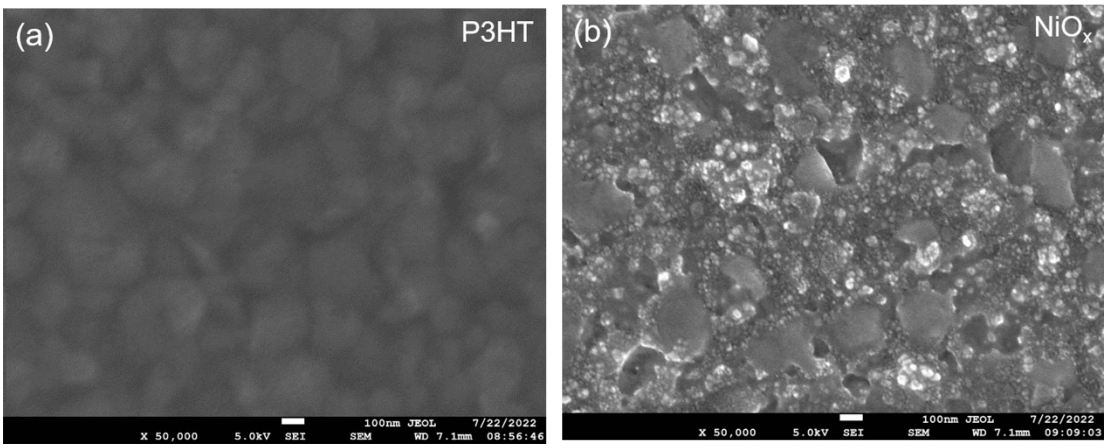

Figure S3. Top-view SEM image of pristine (a) P3HT and (b) NiO_x films.

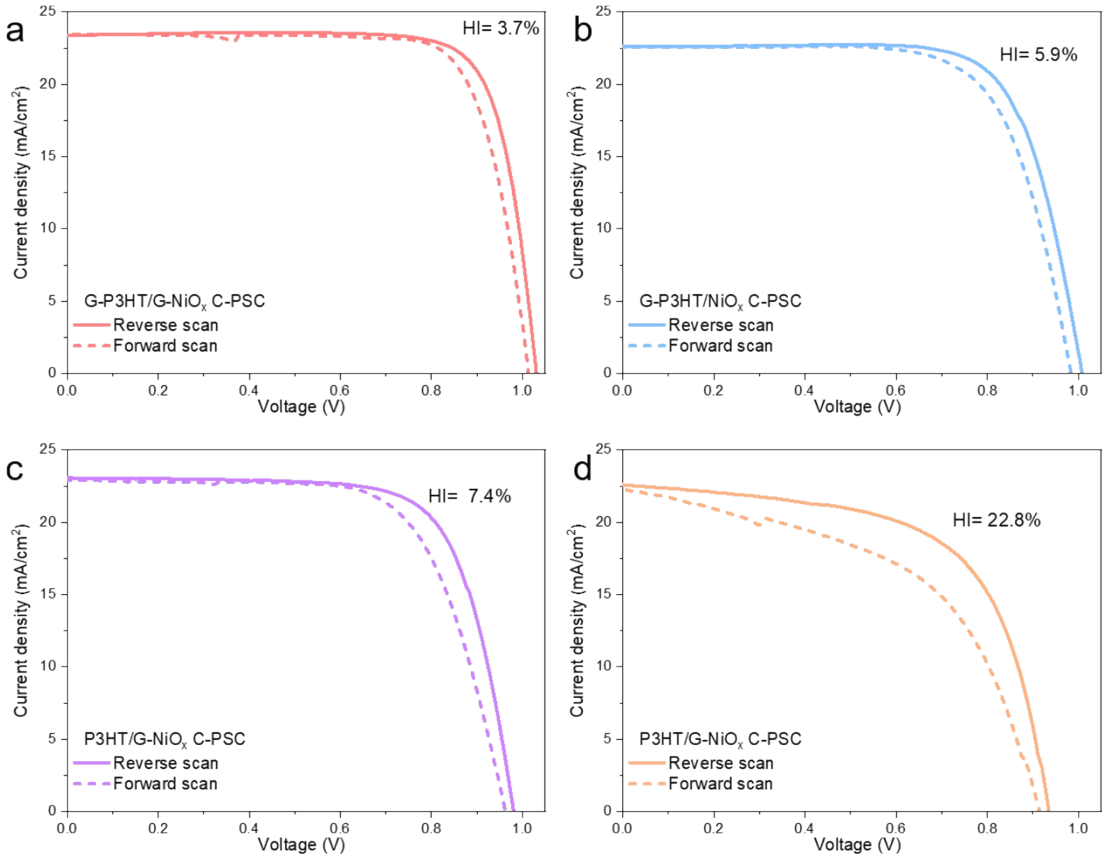

Figure S4. Hysteresis behavior of the C-PSCs with P3HT/NiO_x, P3HT/G-NiO_x, G-P3HT/NiO_x, and G-P3HT/G-NiO_x·bilayers·HTLs.

Table S1. Fitting parameters of the TRPL spectra for the perovskite films modified with P3HT/NiO_x and G-P3HT/G-NiO_x bilayer HTLs.

Samples	A_1 (%)	τ_1(ns)	A_2 (%)	τ_2 (ns)	$\tau_{average}$ (ns)
Perovskite	49.78	51.30	146.14	197.43	185.54
Perovskite/P3HT/NiO_x	2.18	0.82	63.11	114.59	114.56
Perovskite/G-P3HT/G-NiO_x	9.05	0.75	19.79	44.47	44.13