Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Plasma-Assisted Atomic Layer Deposition of Nickel Oxide as Hole Transport Layer for Hybrid Perovskite Solar Cells

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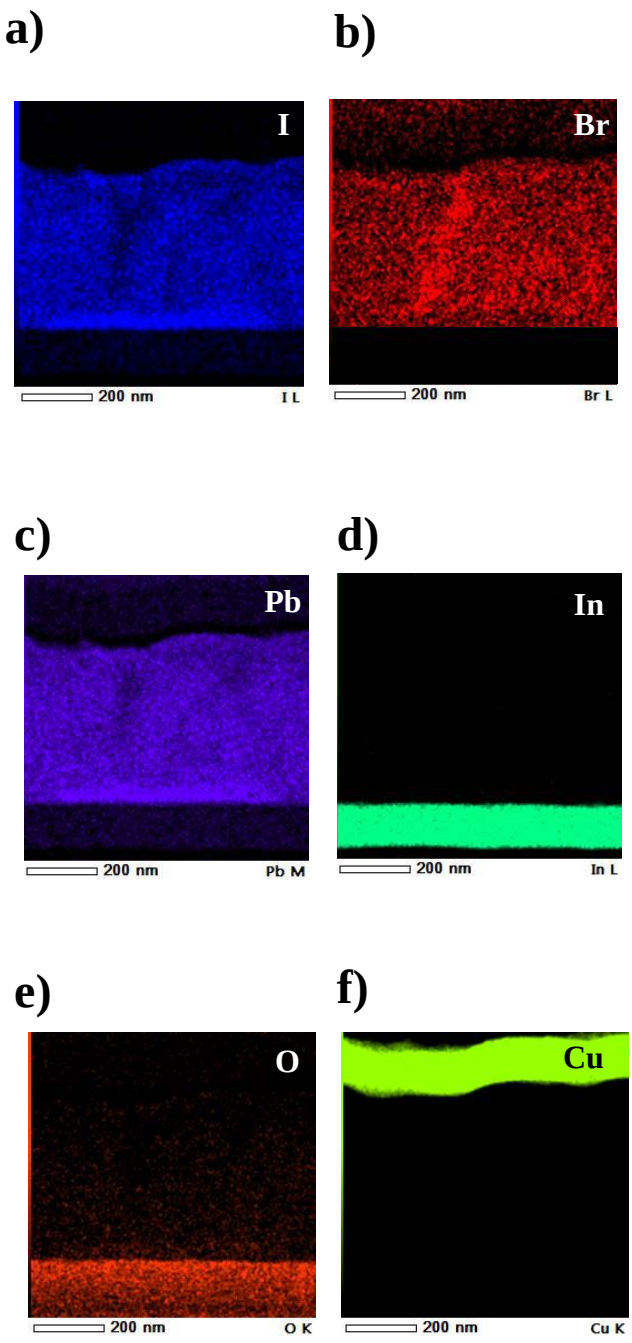

Figure S1. (a-f) TEM-EDX elemental maps of I, Br, Pb, In, O and Cu present in the PSC layer stack.

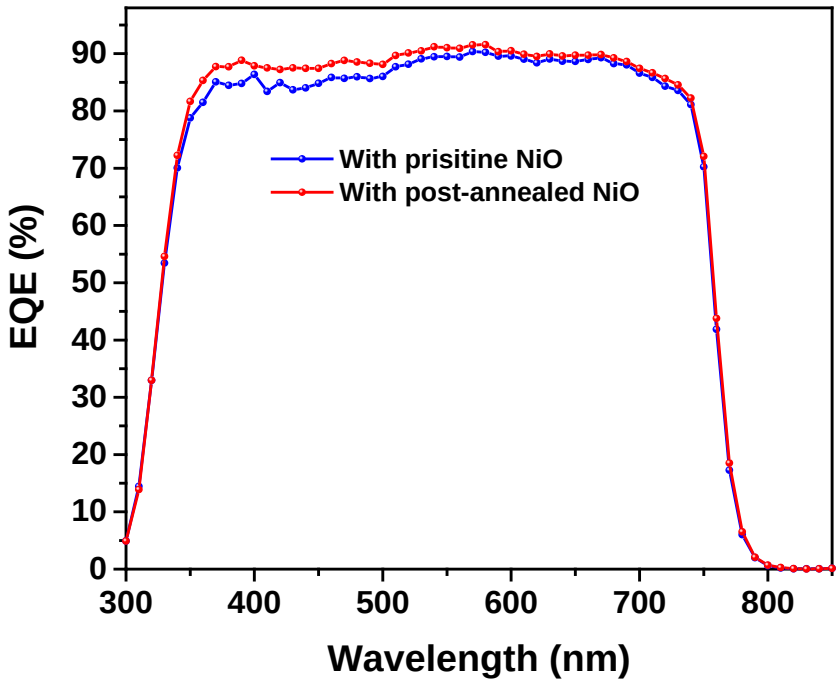

Figure S2. Comparative EQE spectra of PSCs with pristine and post-annealed NiO.

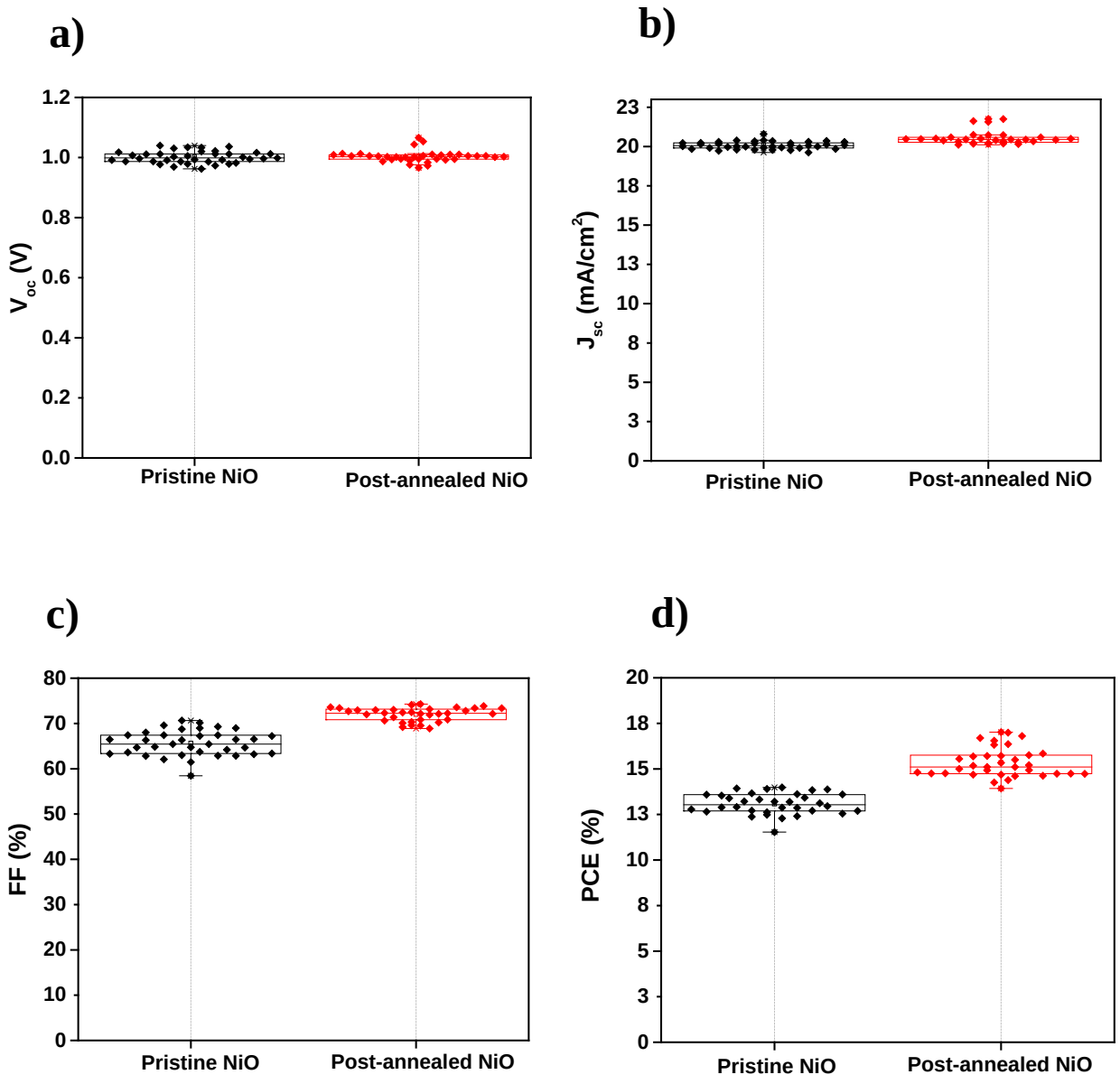

Figure S3. Box plot showing the PV parameters of the PSCs employing pristine NiO and post-annealed NiO (at 300 °C for 20 min in air).

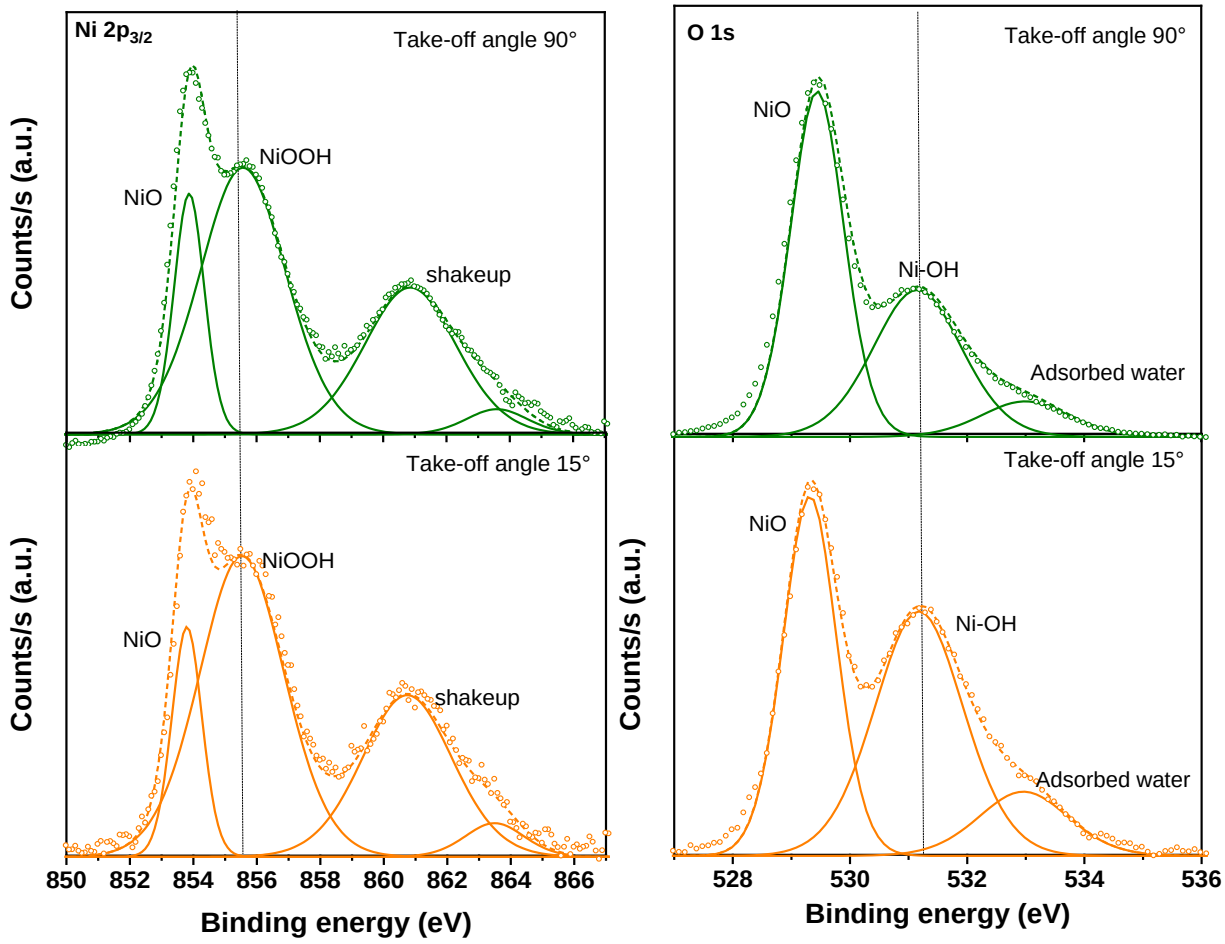

Figure S4. AR-XPS spectra of (a) Ni $2p_{3/2}$ and (b) O 1s of the pristine NiO film measured with photoelectron take-off angles (θ) of 90° and 15° to show that the concentration of hydroxyl species is higher at the surface. Open circles, solid lines and dashed lines are measured data, peak fits and cumulative fits, respectively.

Data Analysis of UPS Spectrum

The typical UPS spectrum is presented in **Figure S5**. As it can be seen, the spectrum has on-set and cut-off edges. The former one is formed by the electrons, which have the weakest binding energy (valence band maximum). The cut-off or secondary electron edge provides information about the slowest secondary electrons which are emitted with zero kinetic energy. A negative bias is applied to the sample to enable detection of these electrons.

For solid samples, the on-set and cut-off energies are found by extrapolating the tangent through the point of inflection of the leading edges of the UPS spectrum to the background level. Once the cut-off and on-set energies are determined (shown in the energy band diagram presented in **Figure S5**), the work function (WF) and ionization energy (IE) can be calculated using the formulas:

$IE = h\upsilon - (E_{cut\text{-}off} - E_{on\text{-}set})$

$WF = h\upsilon - (E_{cut\text{-}off} - E_{Fermi})$

where, $h\upsilon$ is the photon energy (21.22 eV used in this work). The difference between the IE and WF provides information about the position of valence band maximum (VBM) with respect to Fermi level. The position of the Fermi level is determined separately using a clean gold sample.

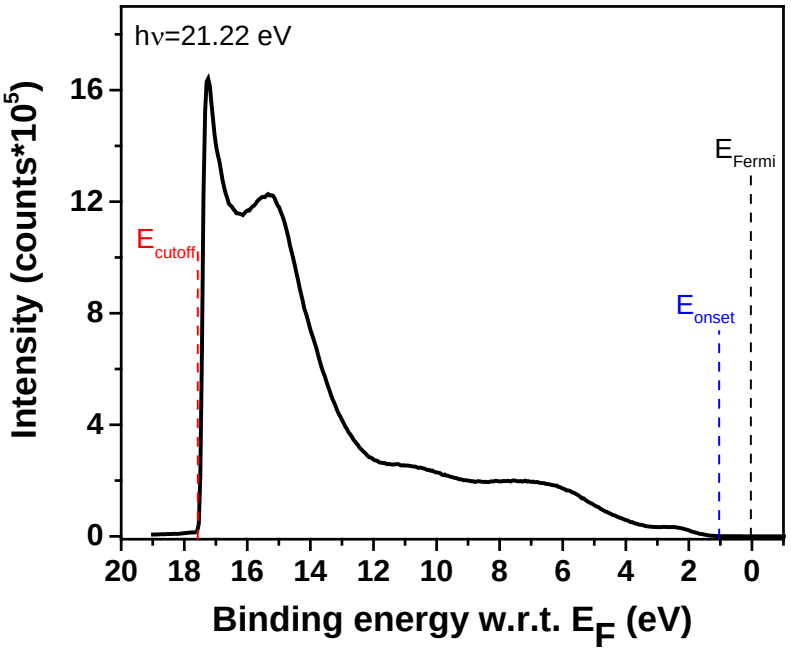

Figure S5. Typical UPS spectrum.

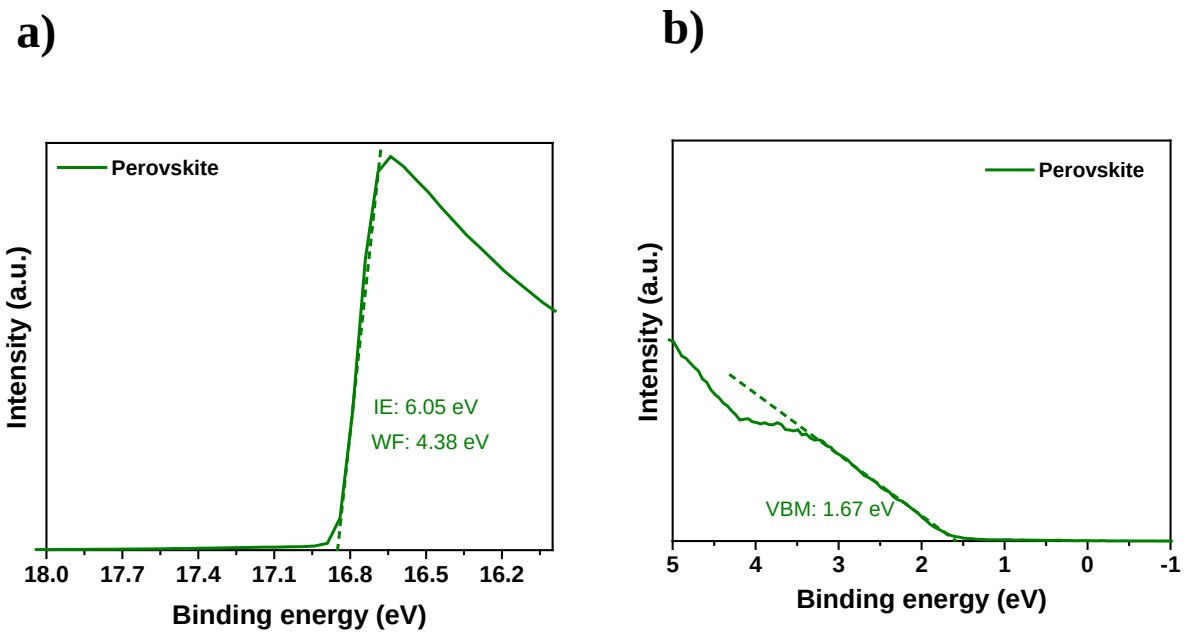

Figure S6. UPS spectra of the employed triple cation-based perovskite absorber layer.

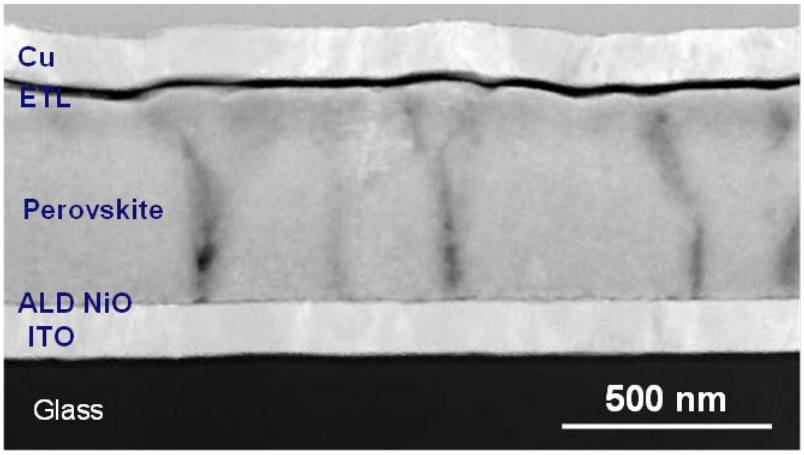

Figure S7. Cross-sectional high angle annular dark field (HAADF) scanning TEM image of the PSC with post-annealed ALD NiO.

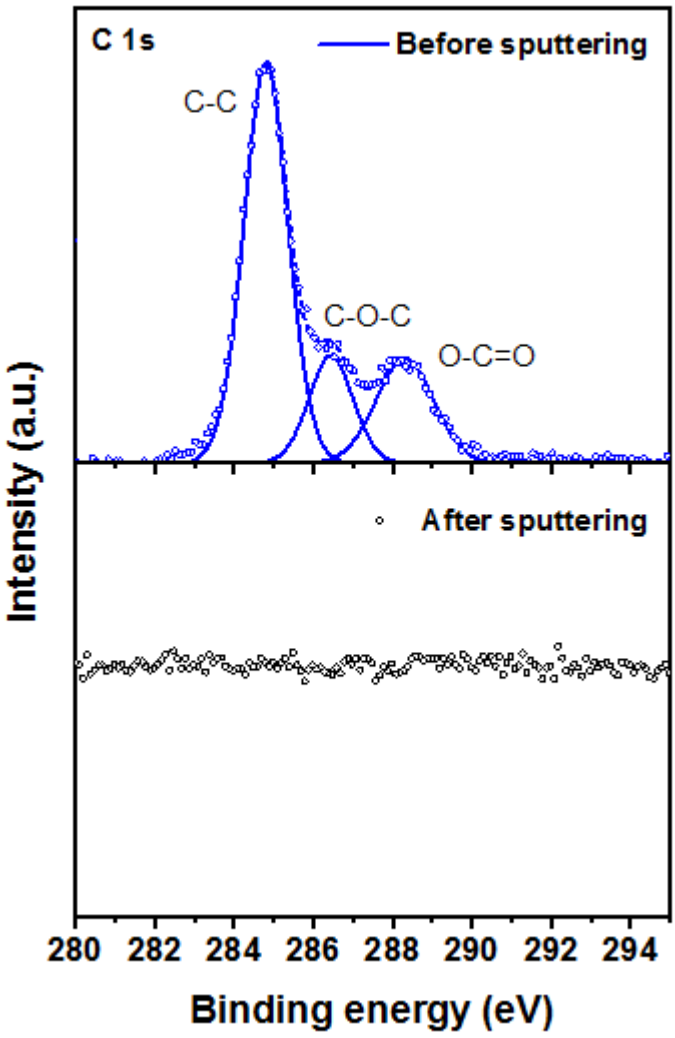

Figure S8. XPS spectra of C 1s of ALD NiO film (a) before and (b) after 10.3 seconds of sputtering with Ar^+ ions. The spectra indicate that C contamination is present only at the surface of the NiO film and not in its bulk. Open circles, solid lines and dashed lines are measured data, peak fits and cumulative fits, respectively.

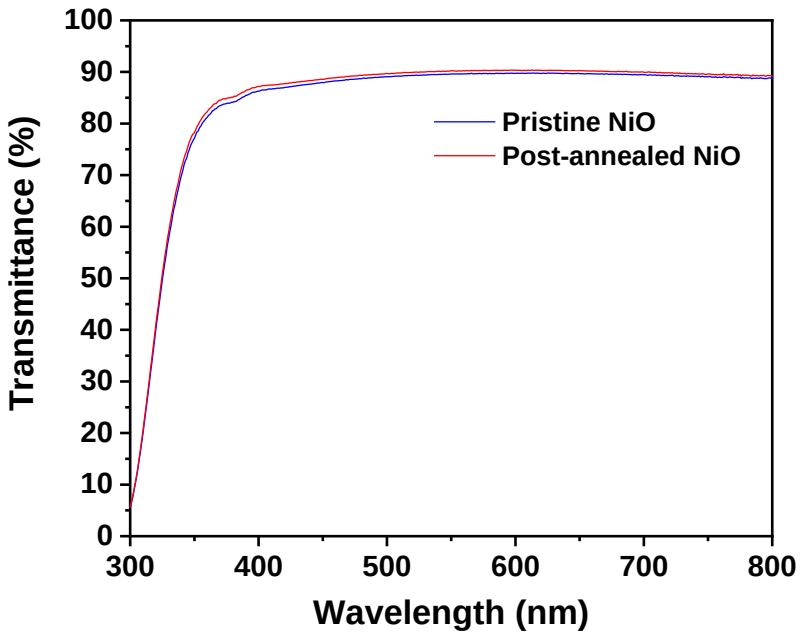

Figure S9. Transmittance spectra of the pristine and post-annealed ALD NiO films.