Impact of Moisture on Efficiency-Determining Electronic Processes in Perovskite Solar Cells Supporting Information

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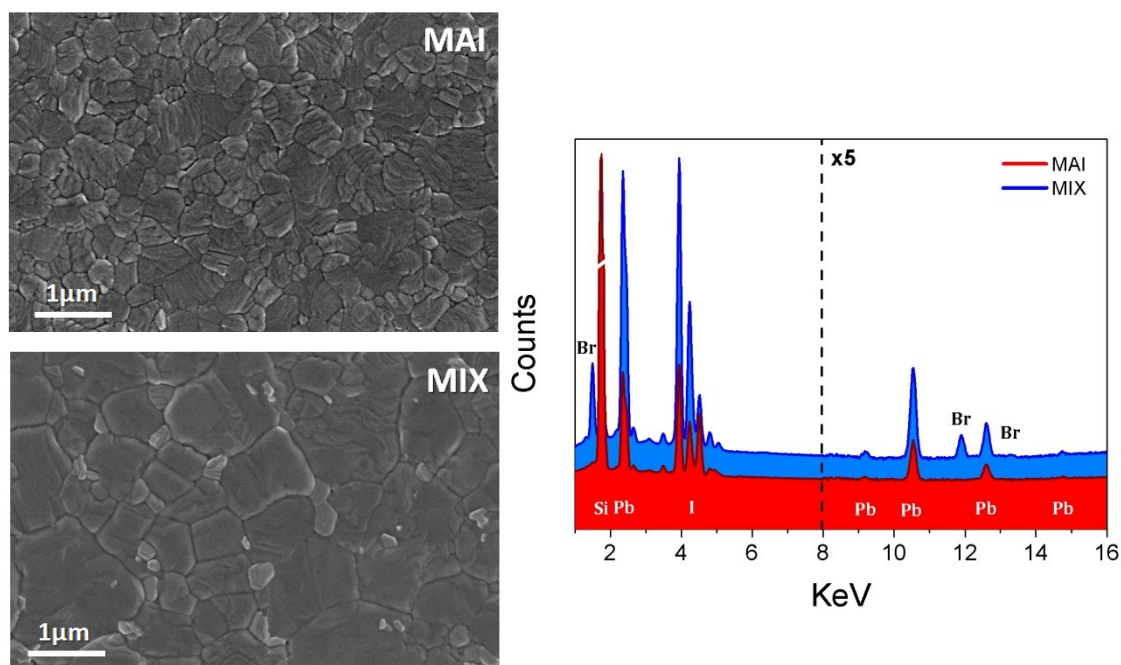
Figure S1. (Left) Plane-view scanning electron microscopy (SEM) images for the (MAI) MAPbI$_3$ and (MIX) Cs$_{0.05}$(MAPbBr$_3$)$_{0.15}$(FAPbI$_3$)$_{0.85}$ perovskite layers. (Right) EDX spectra of the different perovskite layers deposited on Si/cTiO$_2$/mTiO$_2$.
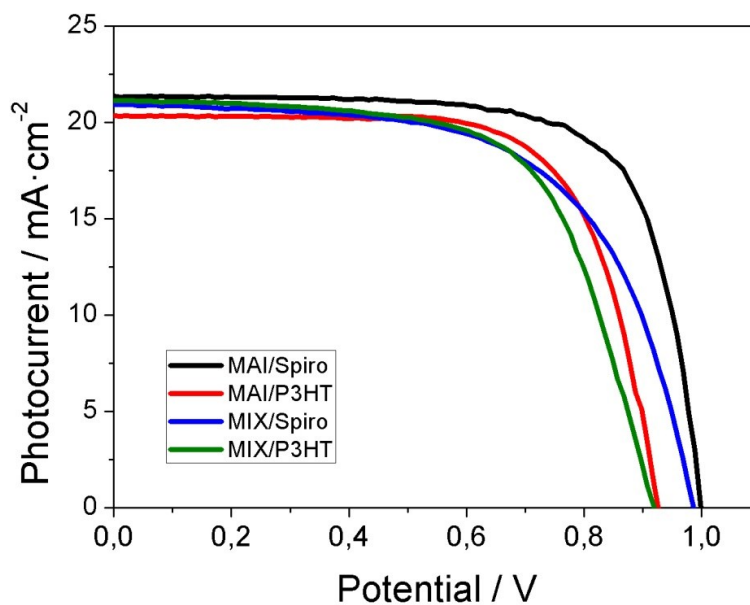
Figure S2. Current-voltage characteristics of the different devices under 1 sun - AM1.5 illumination in reverse scan. A scan rate of 100mV/s with a poling of 30s at 1.2V was employed.
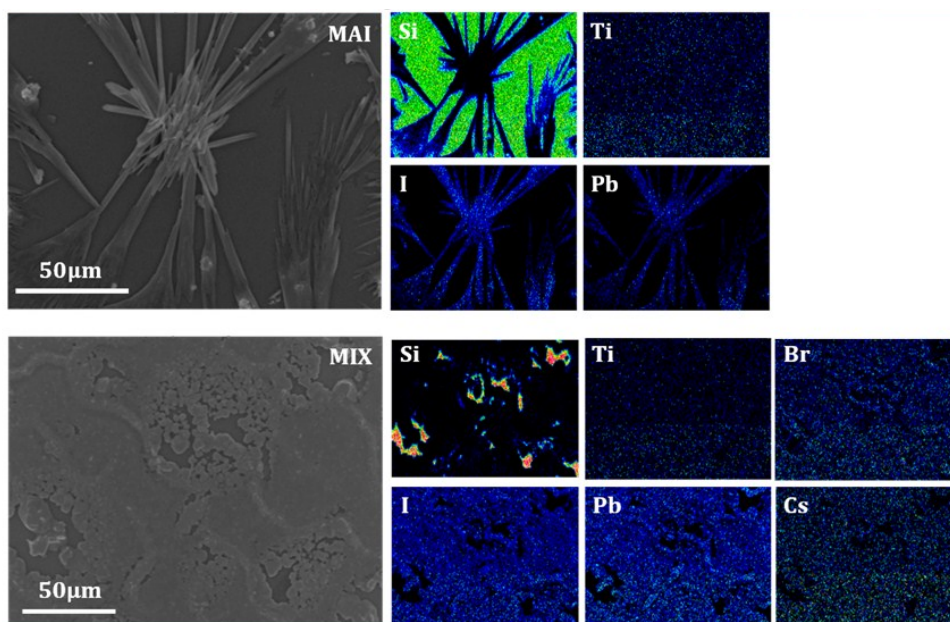

Figure S3. EDX mapping of the different (MAI and MIX) perovskite layers deposited on Si/cTiO$_2$/mTiO$_2$ after moisture exposure (RH > 90%) for 5 hour at room temperature.
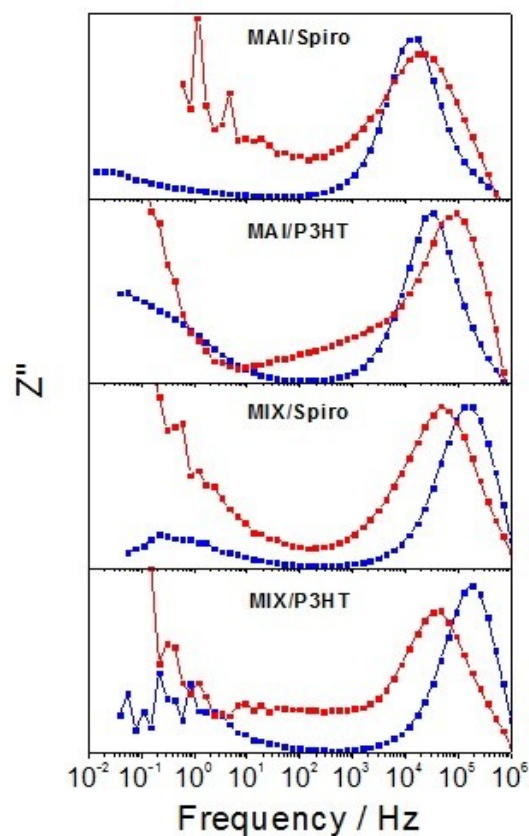
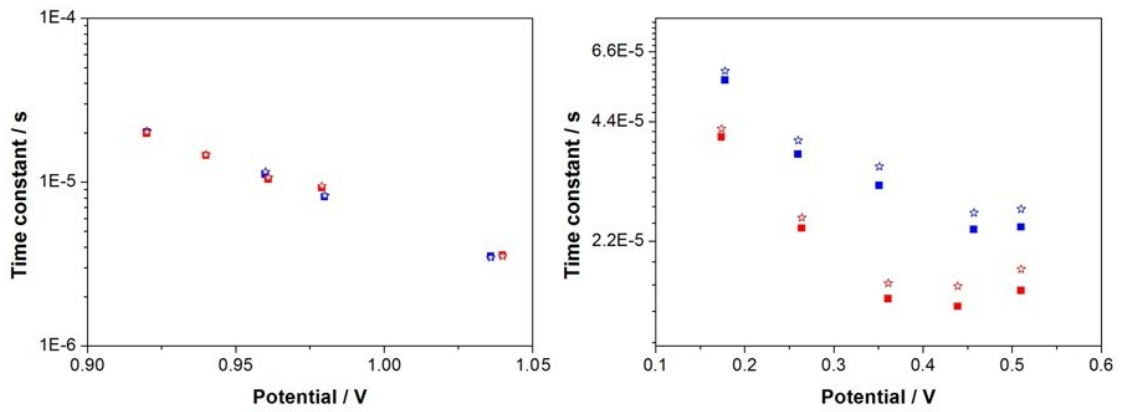**Figure S4.** Impedance frequency plots of the imaginary part for (blue) fresh and (red) degraded PSCs obtained at open-circuit condition and under white illumination.
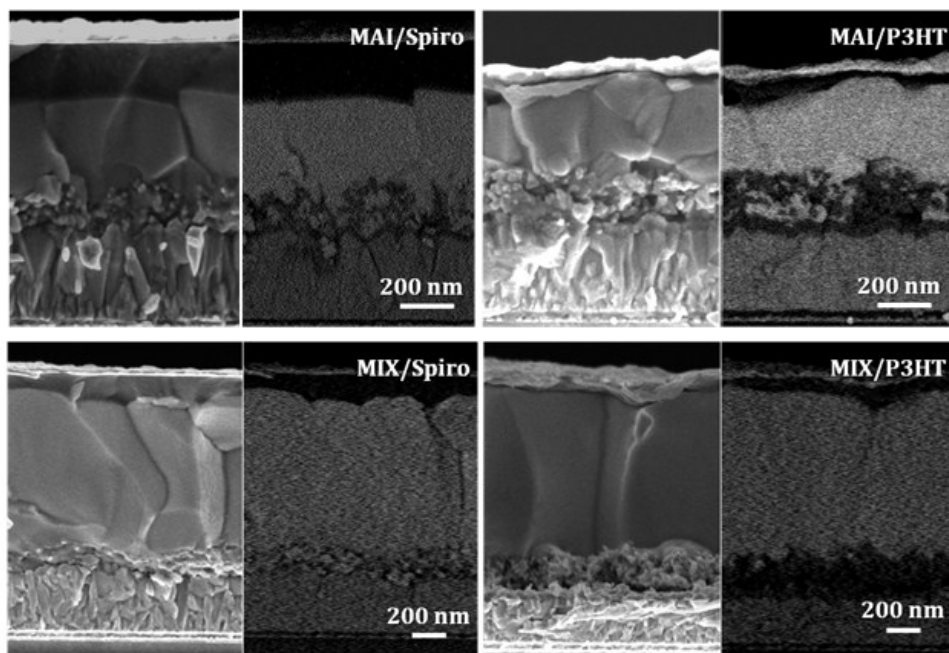
Figure S5. (Right) cross-section scanning electron microscopy and (left) back scattering electron images for the different configurations. A thickness of 450 nm and 950 nm were found for MAI and MIX devices, respectively.
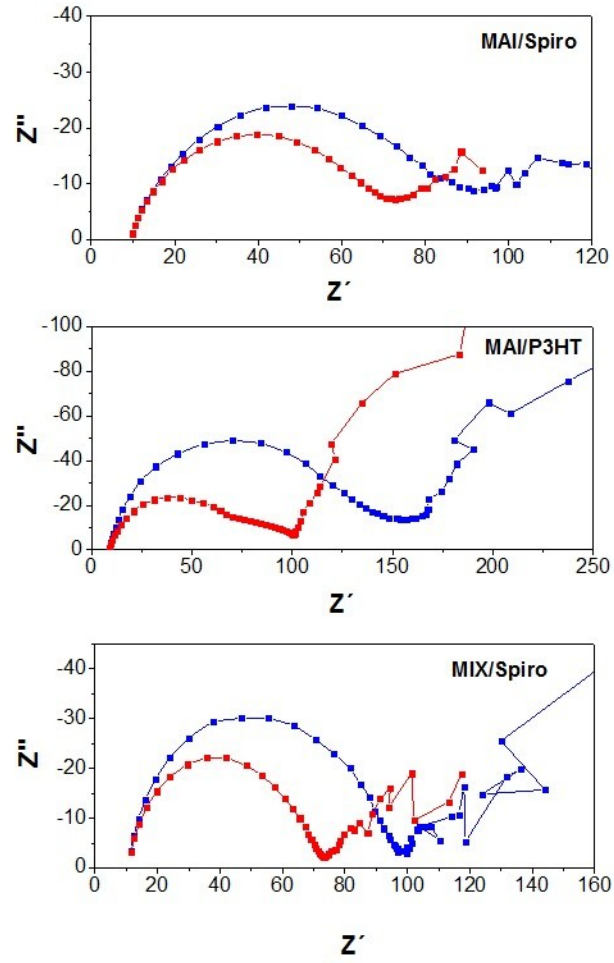
Figure S6. Nyquist impedance spectra for degraded MAI/Spiro, MAI/P3HT and MIX/Spiro devices at open-circuit condition and under blue and red illumination.

Figure S7. HF time constant extracted as (squares) $R_{rec} \cdot C_b$ and (stars) $1/2\pi f$ from impedance response for (right) fresh and (left) degraded MAI/Spiro devices at open-circuit condition and under blue and red illumination.

Figure S8. Electron recombination resistance versus open-circuit potential and under white-light excitation extracted from fittings of the impedance spectra for the (left) fresh and (right) degraded PSCs

Figure S9. Impedance frequency plots of the imaginary part for (left) fresh and (right) degraded PSCs obtained at open-circuit condition and under illumination using the two excitation wavelengths of λ_{blue} = 465 nm (blue) and λ_{red} = 635 nm (red).
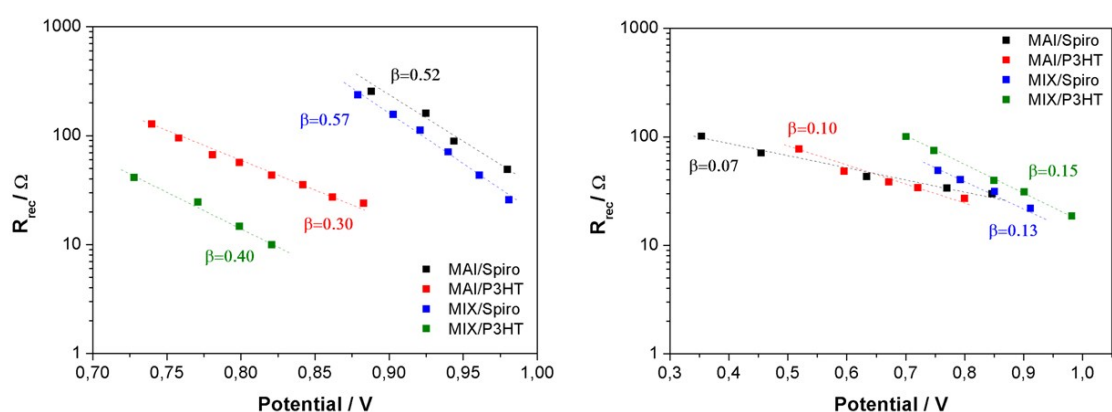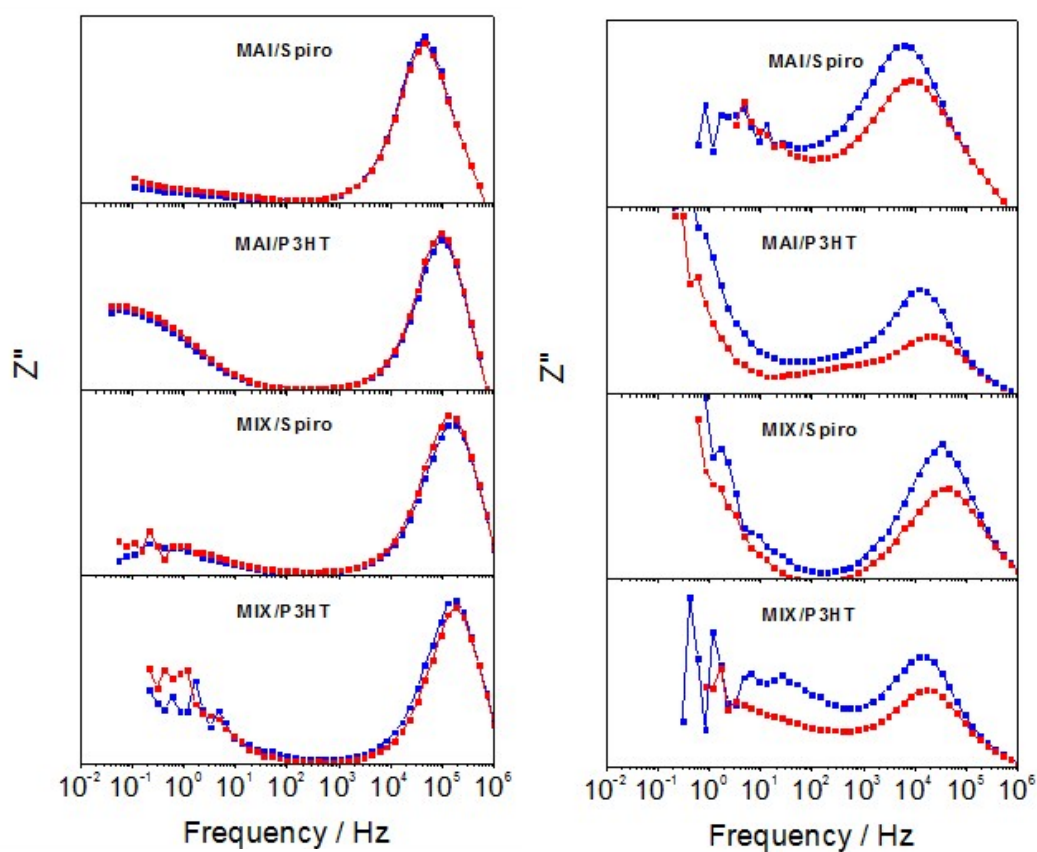
Table S1. Photovoltaic parameters measured under 1 sun - AM 1.5 illumination for the different devices in reverse scan. Error bars are estimated from the results of three best devices of the same configuration.

	J_{SC} (mA·cm^{-2})	V_{OC} (mV)	Fill Factor (%)	Efficiency (%)
MAI/Spiro	20.9 ± 0.2	996 ± 5	73 ± 2	15.3 ± 0.9
MAI/P3HT	20.4 ± 0.3	940 ± 10	70 ± 5	13.2 ± 0.8
MIX/Spiro	21.2 ± 0.1	986± 4	64 ± 3	13 ± 1
MIX/P3HT	21.1 ± 0.2	912 ± 17	65 ± 1	12.5 ± 0.2

Table S2. Static water contact angle of the different devices.

	Water Contact Angle
MAI	72.5 ± 2
MAI/Spiro	77 ± 2
MAI/P3HT	106 ± 3
MIX	87 ± 2
MIX/Spiro	78 ± 2
MIX/P3HT	105 ± 3

Table S3. Atomic percentage of the elements present in the samples estimated after XPS characterization.

Device / At %	Pb	I	O	C	N	S	F	Ti
MAI	15.9	18.6	11.2	47.0	7.3	-	-	-
MAI (90% RH)	7.7	14.8	32.8	27.4	3.4	-	-	13.9
MAI/Spiro	-	-	14.1	73.2	4.5	2.4	5.8	-
MAI/Spiro (90% RH)	0.2	0.7	15.1	68.2	5.6	2.2	8.1	-
MAI/P3HT	-	-	-	88.8	-	11.2	-	-
MAI/P3HT (90% RH)	-	0.4	2.1	87.7	-	9.8	-	-
MIX	13.4	22.8	10.8	44.9	8.1	-	-	-
MIX (90% RH)	14.4	37.9	8.6	28.9	10.2	-	-	-
MIX/Spiro	-	-	12.7	77.2	4.4	2.0	3.6	-
MIX/Spiro (90% RH)	-	0.6	19.1	68.1	4.6	1.7	5.9	-
MIX/P3HT	-	-	-	89.6	-	10.4	-	-
MIX/P3HT (90% RH)	-	-	-	90.7	-	9.3	-	-