Supporting Information

The Role of Surface Energy Control in Organic Photovoltaics Based on Solar Paints

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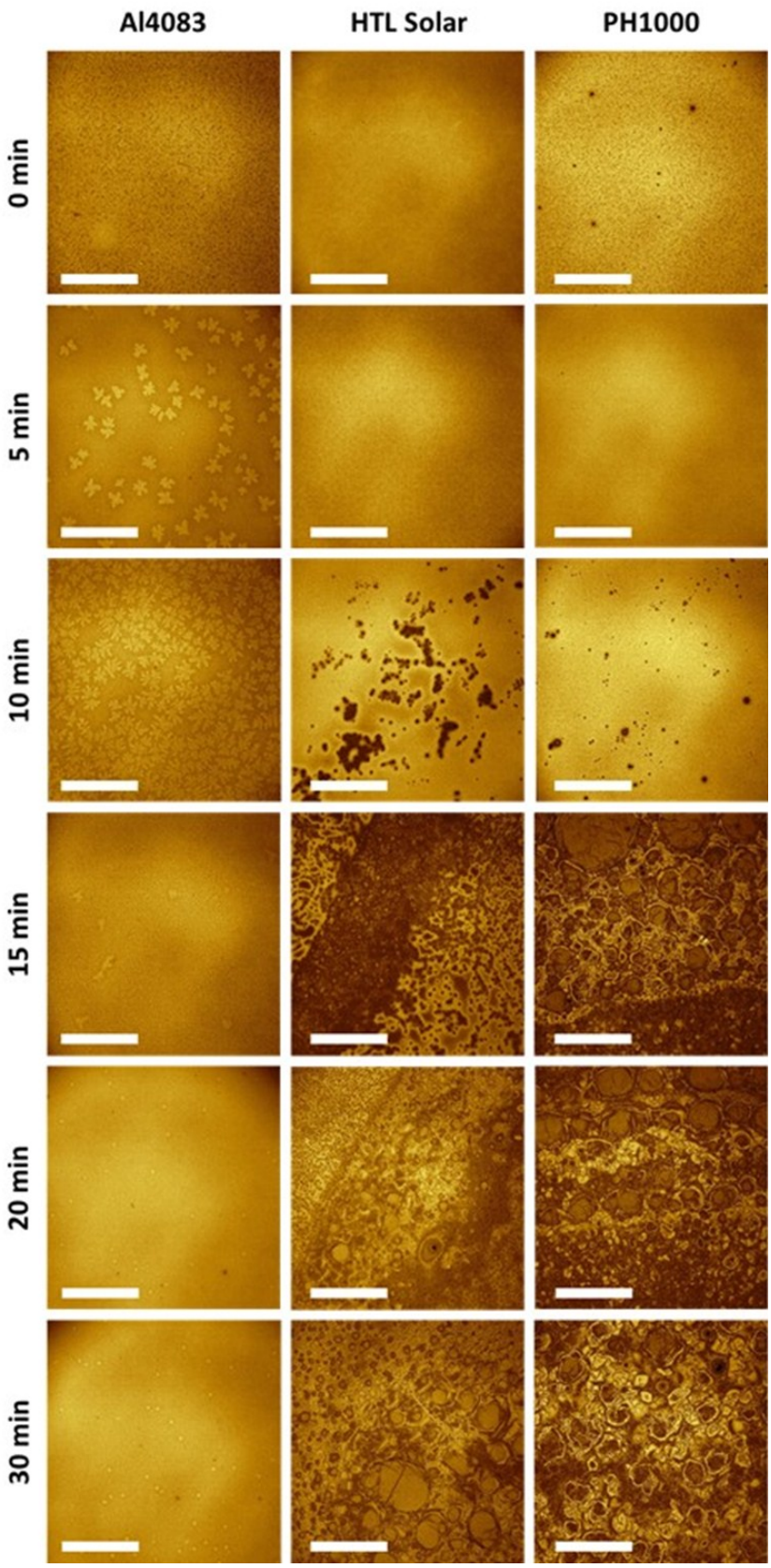


Figure S1. Reflection optical images of nanoparticle films spin coated onto three PEDOT:PSS formulations (Al4083, HTL Solar and PH1000) exposed to UVO for different lengths of time (0 - 30 min). Scale bar is 100 μm.

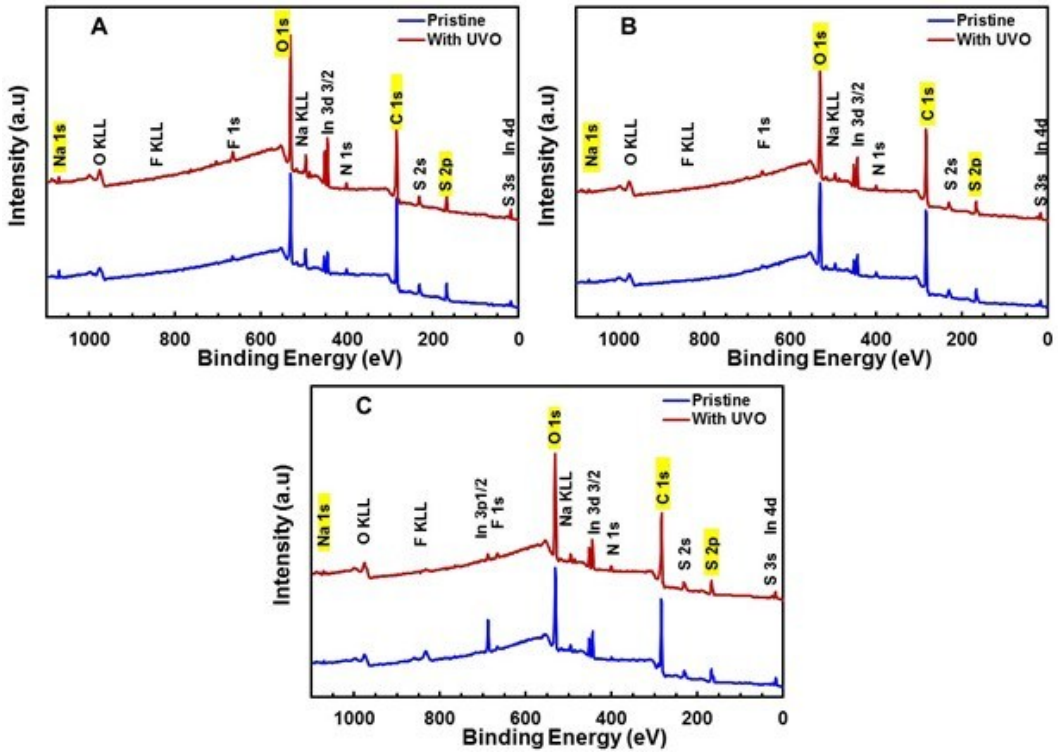


Figure S2. XPS survey spectra of PEDOT:PSS formulations (A) Al4083, (B) HTL Solar, and (C) PH1000 both with and without UVO treatment. Primary carbon, oxygen, sulphur and sodium peaks are labelled.

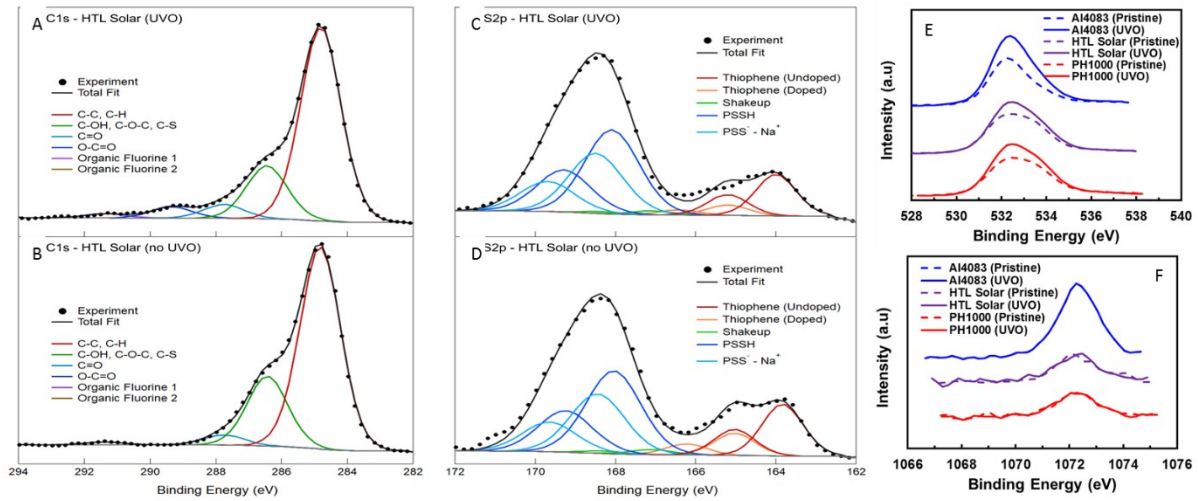


Figure S3. Fitting model applied to pristine HTL Solar S2P peak. C 1s (A, B), S 2p (C, D), O 1s (E) and Na 1s (F) region scans for both the pristine and ozone treated HTL Solar PEDOT:PSS films.

Table S 1: Extracted atomic concentrations from the XPS sulphur regional scan of the three PEDOT:PSS's before and after UVO treatment using the fitting showed in **Figure S3**.

Sample Identifier	Name	Position	FWHM	%At Conc

Al4083 No UVO	Thiophene 3/2 (Undoped)	164.0	1.2	4.4
	Thiophene 1/2 (Undoped)	165.2	1.2	4.3
	Thiophene 3/2 (Doped)	165.2	1.2	3.0
	Thiophene 1/2 (Doped)	166.4	1.2	3.0
	Shakeup 3/2	167.1	1.2	0.4
	Shakeup 1/2	168.3	1.2	0.4
	PSS--Na+ (3/2)	168.1	1.6	24.9
	PSS--Na+ (1/2)	169.3	1.6	24.3
	PSSH (3/2)	168.5	1.6	17.8
	PSSH (1/2)	169.7	1.6	17.4
Al4083 UVO	Thiophene 3/2 (Undoped)	163.8	1.2	3.3
	Thiophene 1/2 (Undoped)	165.0	1.2	3.3
	Thiophene 3/2 (Doped)	165.0	1.2	1.1
	Thiophene 1/2 (Doped)	166.2	1.2	1.1
	Shakeup 3/2	167.1	1.2	0.3
	Shakeup 1/2	168.3	1.2	0.3
	PSS--Na+ (3/2)	168.2	1.6	26.7
	PSS--Na+ (1/2)	169.4	1.6	26.1
	PSSH (3/2)	168.6	1.6	19.1
	PSSH (1/2)	169.8	1.6	18.6
HTL Solar No UVO	Thiophene 3/2 (Undoped)	163.8	1.2	9.8
	Thiophene 1/2 (Undoped)	165.0	1.2	9.5
	Thiophene 3/2 (Doped)	165.0	1.2	4.2
	Thiophene 1/2 (Doped)	166.2	1.2	4.1
	Shakeup 3/2	167.1	1.2	1.0
	Shakeup 1/2	168.3	1.2	1.0
	PSS--Na+ (3/2)	168.0	1.6	20.8
	PSS--Na+ (1/2)	169.2	1.6	20.4
	PSSH (3/2)	168.4	1.6	14.9
	PSSH (1/2)	169.6	1.6	14.5
HTL Solar UVO	Thiophene 3/2 (Undoped)	164.0	1.2	8.5
	Thiophene 1/2 (Undoped)	165.2	1.2	8.3
	Thiophene 3/2 (Doped)	165.2	1.2	2.1
	Thiophene 1/2 (Doped)	166.4	1.2	2.1
	Shakeup 3/2	167.1	1.2	0.8
	Shakeup 1/2	168.3	1.2	0.8
	PSS--Na+ (3/2)	168.1	1.6	22.8
	PSS--Na+ (1/2)	169.3	1.6	22.4
	PSSH (3/2)	168.5	1.6	16.3
	PSSH (1/2)	169.7	1.6	16.0
PH1000 No UVO	Thiophene 3/2 (Undoped)	164.0	1.2	10.6
	Thiophene 1/2 (Undoped)	165.2	1.2	10.4
	Thiophene 3/2 (Doped)	165.2	1.6	4.6
	Thiophene 1/2 (Doped)	166.4	1.6	4.5
	Shakeup 3/2	167.1	1.6	1.1
	Shakeup 1/2	168.3	1.6	1.0
	PSS--Na+ (3/2)	168.1	1.6	20.0
	PSS--Na+ (1/2)	169.3	1.6	19.6
	PSSH (3/2)	168.5	1.6	14.3
	PSSH (1/2)	169.7	1.6	14.0

PH1000 UVO	Thiophene 3/2 (Undoped)	164.0	1.2	9.5
	Thiophene 1/2 (Undoped)	165.2	1.2	9.3
	Thiophene 3/2 (Doped)	165.2	1.2	3.9
	Thiophene 1/2 (Doped)	166.4	1.2	3.8
	Shakeup 3/2	167.1	1.2	1.0
	Shakeup 1/2	168.3	1.2	0.9
	PSS--Na+ (3/2)	168.1	1.6	21.1
	PSS--Na+ (1/2)	169.3	1.6	20.6
	PSSH (3/2)	168.5	1.6	15.1
	PSSH (1/2)	169.7	1.6	14.7

Table S 2: Extracted data from XPS of the carbon regional scan of HTL Solar before and after UVO using the fitting model shown in **Figure S3**.

Sample Identifier	Chemical Assignment	Position (eV)	FWHM	% Atomic Concentration
No UVO	C-C, C=C, C-S	285.0	1.4	68.9
	C-OH, C-O-C	286.5	1.4	23.8
	C-CF, C=O	287.7	1.4	5.6
	C-F	288.5	1.4	0.0
	O-C=O	289.3	1.4	0.1
	pi-pi*	291.5	2.0	1.6
UVO	C-C, C=C, C-S	285.0	1.4	69.6
	C-OH, C-O-C	286.6	1.4	18.6
	C-CF, C=O	287.7	1.4	5.7
	C-F	288.5	1.4	0.0
	O-C=O	289.3	1.4	4.0
	pi-pi*	291.1	2.1	2.2

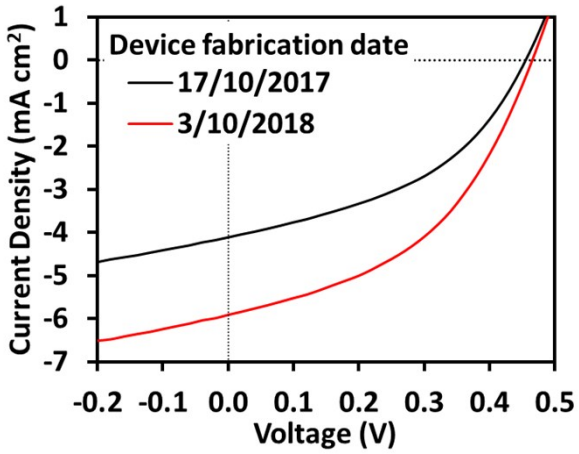


Figure S4. Test of inks stability as a function of time, these devices are prepared from the same batch of ink (prepared on 2/9/17) but with a year apart.

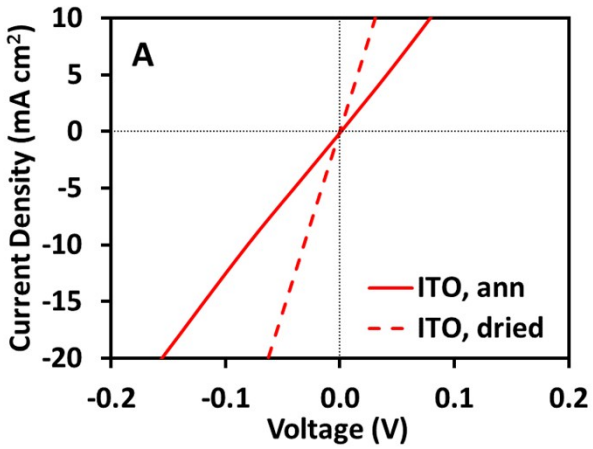
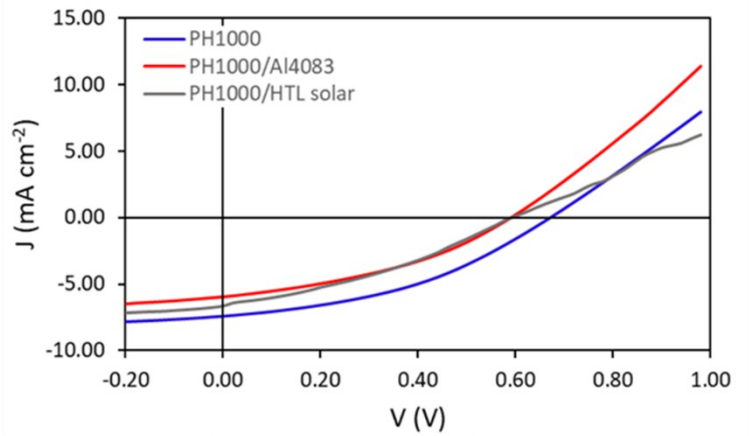


Figure S5. Testing the importance of the PEDOT:PSS layer for NP-OPV. The devices presented here has been prepared with the geometry glass/ITO/NP-active layer/Al, whereas this geometry work for BHJ devices it does not prepare working devices for NP-OPV.



Bottom electrode	PCE (%)	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (#)
PH1000	1.84 (0.16)	0.63 (0.04)	-7.28 (0.34)	0.40 (0.03)
PH1000/Al4083	1.17 (0.13)	0.61 (0.05)	-5.67 (0.56)	0.34 (0.03)
PH1000/HTL Solar	1.15 (0.19)	0.57 (0.02)	-5.69 (0.80)	0.33 (0.02)

Figure S6. I-V curves of roll-to-roll prepared devices. The devices have been prepared with a bottom electrode of Ag/PH1000 for charge transport, the blue line is the I-V characteristics for a device prepared directly on top of PH1000, the red line has a layer of Al4083 as HTL and the grey line has a layer of HTL Solar as HTL. The device geometry was PET/Ag/PH1000/HTL (if applied)/Active layer/ZnO/Al (Sputtered)