

Supplemental information

Reducing burn-in voltage loss in polymer solar cells by increasing the polymer crystallinity

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Active layer heating under illumination and associated reversible V_{oc} losses

Changes in the temperature of the active layer of organic solar cells upon illumination cause a reversible drop of the open circuit voltage as the solar cell heats under illumination (Figure S1). The active layer temperature is hard to measure directly, but the V_{oc} changes can be used as a proxy, since the open circuit voltage of a solar cell depends linearly on the temperature.^[1] We measure a slope of 0.9 meV/K for a PCDTBT solar cell. Using this dependence we could calculate a temperature difference in the active layer of about 12 °C for halogen and 2.5 °C for LED illumination after two minutes of illumination. Notably, this temperature increase in the active layer happens at a controlled, constant temperature of the glass substrate. The associated V_{oc} losses are fully reversible upon cooling of the cell, as seen in the figure below.

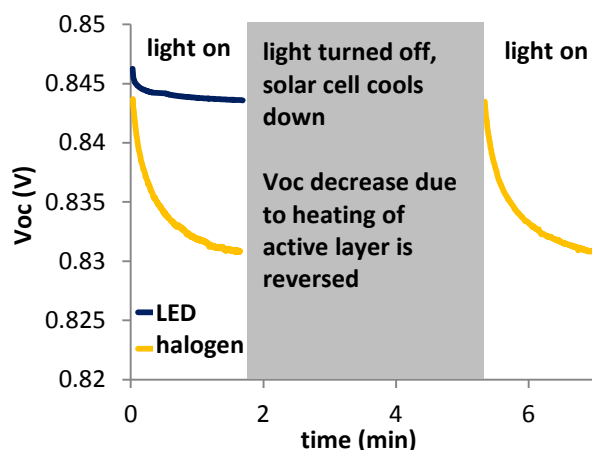


Figure S1. Reversible open circuit losses due to heating of the active layer in temperature controlled substrates. A strong V_{oc} loss due to heating can be observed under halogen illumination, although the substrate temperature is constant. After a cooling period of ca. 3 minutes in the dark, the V_{oc} loss is restored and starts to decrease again. When using LED illumination heating of the active layer is minimized.

Since permanent degradation is fastest during the first minutes, a clear distinction between reversible and irreversible degradation is often not possible from the curves. Thus it is crucial to minimize the temperature changes due to illumination. For a direct comparison IV curves before and after the burn-in experiment provide a temperature independent measurement.

Especially for low T_g polymers like P3HT, a temperature increase of the active layer under illumination could cause the cell to surpass T_g which would activate thermal degradation mechanisms. Since we only study light-induced degradation, a proper estimation and control of the active layer temperature was necessary.

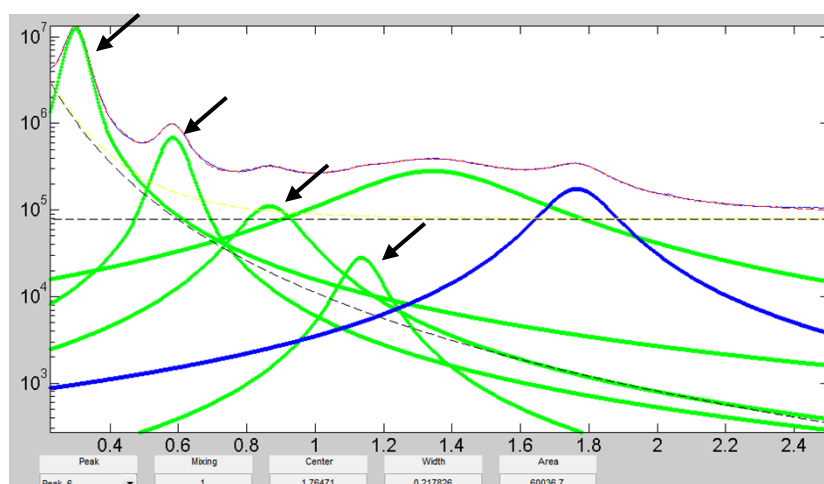


Figure S2. Four distinguishable orders of the lamellar peak in KP115 indicate a high crystallinity. The line plot shows the integrated intensity between $\chi = 60^\circ \dots 120^\circ$ with fits to the four lamellar peaks. The π - π -stacking peak is shown in blue.

Table S1. Device parameters of fresh solar cells from amorphous (PCDTBT, RRa P3HT) and crystalline (P3HT, KP115) materials

fresh performance	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	efficiency (%)
PCDTBT	-10.4	0.87	60	5.4
RRa P3HT	-4.2	0.91	40	1.5
P3HT	-10.7	0.56	72	4.4
zz115	-13.1	0.62	64	5.2

Table S2. Percentage loss of each solar cell parameter for amorphous (PCDTBT, RRa P3HT) and crystalline (P3HT, KP115) materials

burn in loss	J_{sc}	V_{oc}	FF	efficiency
PCDTBT	1.9 %	3.5 %	12.0 %	16.4 %
RRa P3HT	1.3 %	4.0 %	7.8 %	12.4 %
P3HT	1.9 %	0.0 %	3.3 %	5.5 %
zz115	2.3 %	1.2 %	2.3 %	5.4 %

Artificially introducing traps

To verify the accuracy of our transient photocurrent measurements, we artificially introduced traps into a solar cell by evaporating a C_{60} layer on top of the solution processed P3HT:PCBM layer before electrode deposition. Since C_{60} has a LUMO^[2] that is 0.2eV lower than the LUMO of PCBM^[3], it acts as an electron trap in the fullerene phase. After cell production most of the C_{60} is still inside the evaporated top layer of about 10 nm thickness, and the transient photocurrent signal is rather low. Rapid diffusion of fullerenes in P3HT also at low temperatures has recently been shown Treat et al.^[4] Consequently, upon aging, C_{60} can diffuse into the active layer and acts as trap there. This causes a strong increase of the transient photocurrent signal. Around 100 μ s, where light-induced traps were observed after degradation, the signal has increased by more than one order of magnitude for the cell with C_{60} . This reference measurement shows that the transient photocurrent technique can display changes in the trap density of the active layer. The amount of traps in this experiment can roughly be estimated to about 5%, assuming that the initially 10nm layer of C_{60} is distributed homogenously across the 200nm active layer in the aged solar cell. This suggests that the number of traps formed during ageing in regular solar cells is well below 5%, since the signal observed in the cell with C_{60} is much higher than what is observed after a regular ageing experiment.

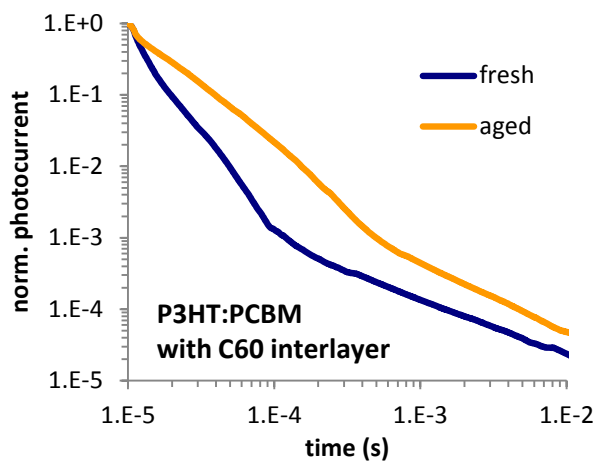


Figure S3. Transient photocurrent measurements for a P3HT:PCBM cell with a 10 nm C_{60} interlayer at the top contact before and after aging. Upon aging the C_{60} diffuses into the active layer and acts as trap there.

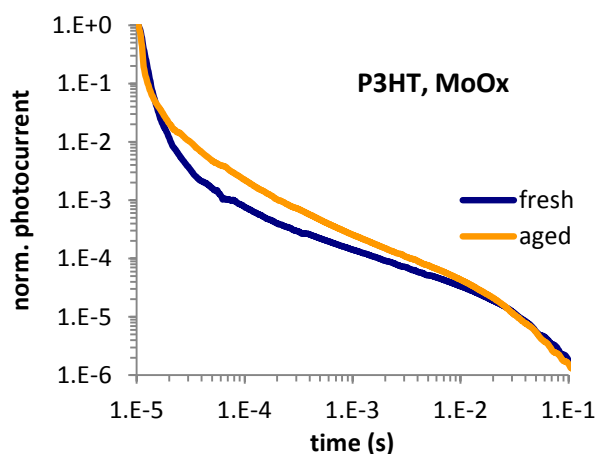


Figure S4. Light-induced trap formation in a solar cell with MoOx bottom contact shows the same behavior as the cell with PEDOT:PSS (Figure 5 in the main text). This shows the independency of our study from the bottom contact.

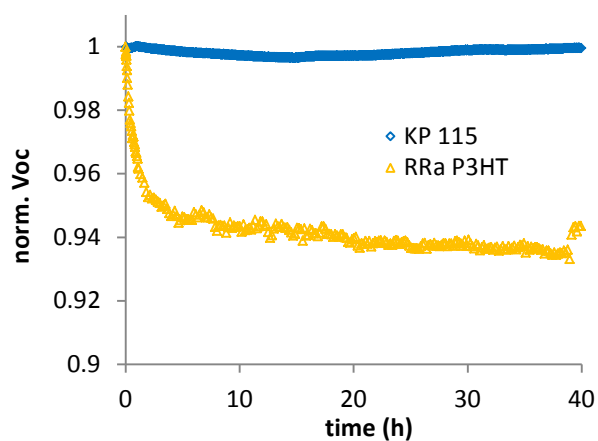


Figure S5. Normalized open circuit voltage losses over illumination time for KP115 and RRa P3HT solar cells.

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