Controlling electrical and optical properties of polymer-fullerene bulk heterojunctions by precisely tuning the degree of polymer aggregation and phase separation from the fullerene

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It is common knowledge that the polymer conformation and its phase separation with fullerene derivatives are delicate issues, crucially impacting on the photovoltaic parameters of polymer based solar cells. Whereas strongly intermixed polymer:fullerene phases provide large interfacial area and consequently a high quantum efficiency of exciton dissociation, pristine and primarily ordered polymer and fullerene domains support efficient charge transport and percolation. To study the aggregation and phase separation behaviour in polymer solar cells we investigated two approaches: firstly the counterbalancing influences of polymer solution concentration and blending ratio with PCBM ([6,6]-phenyl-C61-butyric acid methyl ester) on the basis of a semi-crystalline anthracene-containing poly(*p*-phenylene-ethynylene)-*alt*-poly(*p*-phenylene-vinylene) (PPE-PPV) copolymer statistically bearing either branched 2-ethylhexyloxy or linear octyloxy side-chains (AnE-PV*stat*), and secondly the polymer:polymer blending ratio in ternary blends of a semi-crystalline and an amorphous polymer, bearing the linear octyloxy side-chains at the PPE-part and the branched 2-ethylhexyloxy at the PPV-part (AnE-PV*ab*) and vice versa (AnE-PV*ba*), with PCBM. We demonstrate by carefully choosing solution and PCBM concentration the control of polymer aggregation, and by polymer:polymer blending ratio control of domain size evolution and phase separation. We explicitly demonstrate the counterbalancing effect on charge generation and transport for increasing polymer aggregation and domain purity, in agreement with theoretical considerations. Furthermore the influence of polymer aggregation and phase separation on fundamental optoelectronic properties is discussed, providing detailed understanding of resulting photovoltaic parameters.