SUPPLEMENTARY INFORMATION for

Elasto-morphology of P3HT:PCBM Bulk Heterojunction Organic Solar Cells

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Simulation procedure

We simulate solvent evaporation followed by thermal annealing of the solvent-free morphology. Typical solvent evaporation is simulated under an NPT (pressure constrained at 1 atm. and temperature at 298 K) ensemble by removing 1.25% of the CB molecules from the energy-minimized ternary mixture after every 3 ns (nanosecond) of time interval from a simulation box with 3-dimensional (3D) periodic boundary condition (PBC). Thermal annealing of the solvent-free mixture is achieved by gradually increasing the temperature from 298 K to 698 K over three successive computational steps. This leads to an overall simulation time ~ 3 μs to prepare thermally annealed BHJ composite structures as shown in Figure S1. Simulation details for various cases simulated are presented in Table S1. Considering a mean time-scaling factor ~ 4, the BHJ morphology obtained from CGMD simulations is equivalent to ~ 12 μs of all-atom MD simulation time as also explained elaborately in our previous effort [1]. After achieving solvent-free thermally annealed 3D microstructure, morphological parameters such as domain size, interfacial area to

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volume ratio and percolation ratio are calculated from a spatially discretized structure as shown in Figure S2. Under the assumption of finite element scheme, we discretize the overall simulation box into small voxels of dimensions ~ 0.5×0.5×0.5 nm^3. The dimension of each voxel is limited by the smallest Van der Waals diameter of each CG beads considered.

Table S1: *MD simulation parameters related to energy minimization, equilibration (under NVT and NPT ensembles) and the solvent evaporation simulation are listed. Velocity rescale and Parrinello-Rahman methods are used as thermostat and barostat, respectively, to achieve smooth convergence for temperature and pressure constraints.*

	NVT Equilibration	**NPT Equilibration**	**Solvent Evaporation**	**Thermal Annealing**	**Uniaxial straining (NE-CGMD)**
Algorithm	Leap frog	Leap frog	Leap frog	Leap frog	Leap frog
Time step (*fs*)	20	20	20	20	20
LJ cut-off scheme	Verlet	Verlet	Verlet	Verlet	Verlet
Cut-off radius (*nm*)	1.1	1.1	1.1	1.1	1.1
Thermostat	Berendsen	Berendsen	Velocity-rescale	Velocity-rescale	Velocity-rescale
Barostat	Berendsen	Berendsen	Parrinello-Rahman	Parrinello-Rahman	Parrinello-Rahman
Coupling constant (ps^{-1})	1	2	15	15	15
Temperature (*K*)	298	298	298	298-698	298-698
Pressure (*atm.*)		1	1	1	1 *(transverse)*

	NVT Equilibration	**NPT Equilibration**	**Solvent Evaporation**	**Thermal Annealing**	**Uniaxial straining (NE-CGMD)**
Deformation (nm/ps)					1×10^{-4} *(longitudinal)*

Table S2: *Simulation details for different P3HT:PCBM BHJ active layers considered.*

Weight ratio (P3HT:PCBM)	**Chain length**	**P3HT molecules**	**PCBM molecules**	**Oligomer weight fraction (%)**	**Oligomer molecules**	**Dimension of simulation box (nm^3)**
1.0:0.5	120	119	1306	0	0	20×20×12.110
1.0:0.8	120	119	2090	0	0	20×20×14.335
1.0:1.0	120	119	2612	0	0	20×20×15.732
1.0:2.0	120	119	5225	0	0	20×20×14.360
1.0:0.8	120	119	2213	5	90	20×20×15.258
1.0:0.8	120	119	2319	10	180	20×20×15.957
1.0:0.8	120	119	2424	15	270	20×20×16.726
1.0:0.8	120	119	2529	20	360	20×20×17.441

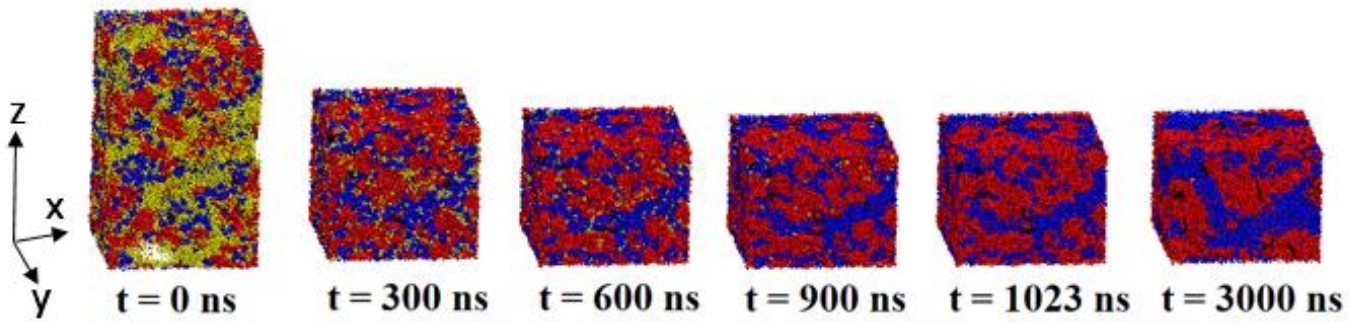

Figure S1: *Simulated evaporation of P3HT:PCBM BHJ active layers from CB solvent at different time steps. More than 50% of CB is observed to evaporate at t = 300 ns. Morphology evaluation and dynamics of evaporation and thermal annealing demonstrate phase-separated BHJ microstructure.*

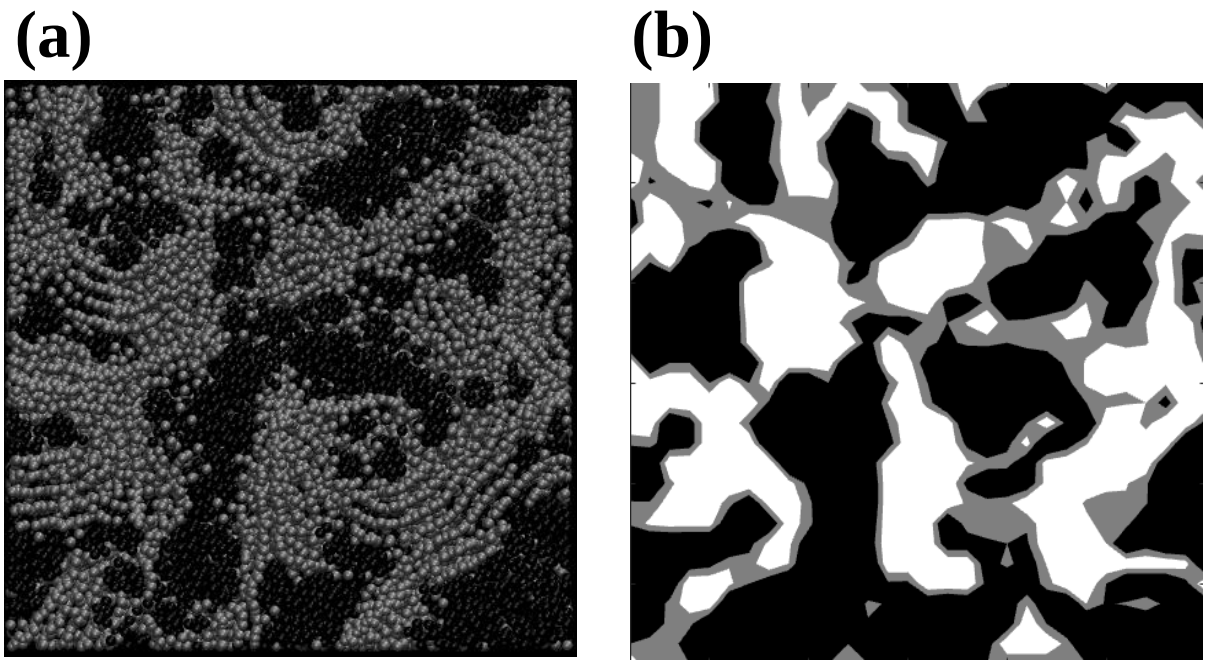

Figure S2: *Comparison of morphologies with spatial discretization (sampled along the x-y plane) for different weight ratios. (a) Snapshots of the BHJ morphology for 1:0.08 weight ratio of P3HT:PCBM. P3HT molecules are represented using grey beads and PCBM molecules are denoted using black beads. (b) The corresponding spatially discretized morphologies for 1:0.08 weight ratio. All the representations are across the x-y plane. White regions represent pure P3HT domains while black represents pure PCBM. Grey areas indicate a mixture of P3HT and PCBM.*

References

S1. Munshi, J., R. Dulal, T. Chien, W. Chen, and G. Balasubramanian, *Solution Processing Dependent Bulk Heterojunction Nanomorphology of P3HT/PCBM Thin Films.* ACS applied materials & interfaces, 2019. **11**(18): p. 17056-17067.