Supplementary Information

Evolution of Molecular Aggregation in Bar-coated Non-fullerene Organic Solar Cells

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1. Fitting of the film thickness using a Cauchy model.

The film thickness (either a wet or dry film) can be are determined using a Cauchy model in the optical transparent range of a thin film, which is based on the principle of the Cauchy model equation (1) and can be expressed in equation (1) below [1]:

$$n(\lambda) = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4} \tag{1}$$

where refractive index (n) is considered as a function of wavelength. A, B and C are parameters which are determined during model fitting. These four parameters (thickness, A, B, C) are employed to calculate Ψ and Δ. Deviations of the fitted and experimental Ψ and Δ values are evaluated via the mean square error (MSE), which is expressed using the following equation (2) [2]:

$$MSE = \frac{1}{2N - M}\sum_{i=1}^{N}\left[\left(\frac{\Psi_i^{mod} - \Psi_i^{exp}}{\sigma_{\Psi, i}^{exp}}\right)^2 + \left(\frac{\Delta_i^{mod} - \Delta_i^{exp}}{\sigma_{\Delta, i}^{exp}}\right)^2\right] \tag{2}$$

where N and M represent the numbers of wavelengths and the number of fit parameters, respectively. The optimized thickness with the correlated parameters (A, B and C) corresponds to the minimum value of MSE.

2. Dynamic measurement raw data of Ψ and Δ.

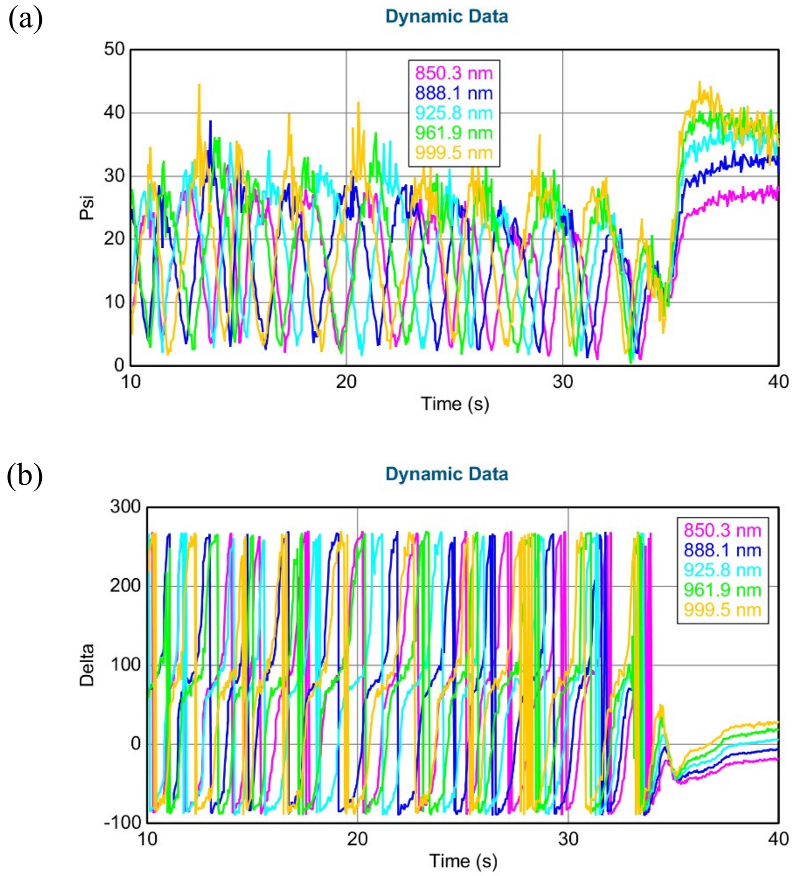
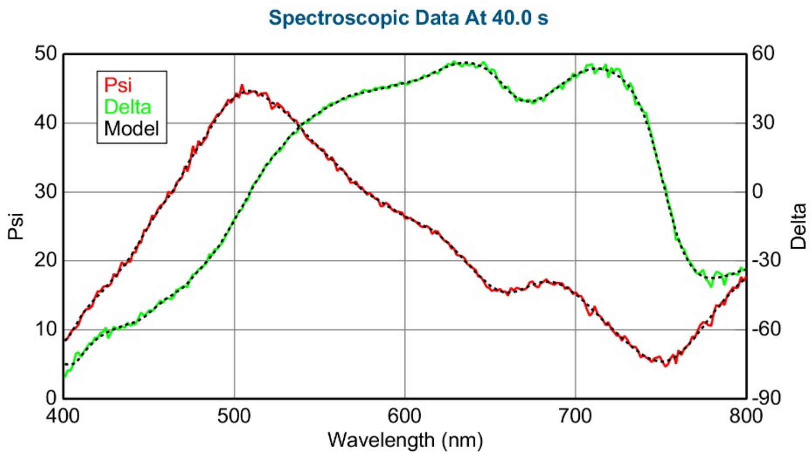


Figure S1. The evolution of SE parameter (a) Ψ and (b) Δ at several selected wavelengths during the PBDB-T:ITIC film drying process.

3. B-spline fitting and optical constants.

(a)



(b)

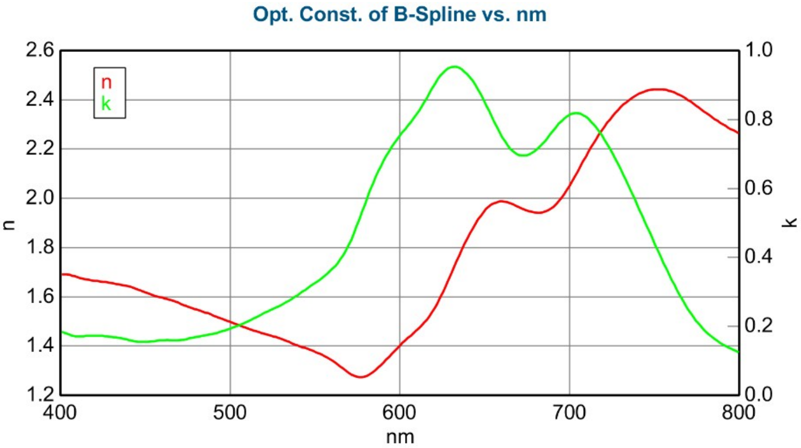


Figure S2. (a) Raw data and the fitted Ψ and Δ (dashed line) using the B-spline model in the wavelength range of 400-800 nm. (b) Extinction coefficient (k) and refractive index (n) obtained via B-spline model fitting.

4. Normalized extinction coefficient in stage IV.

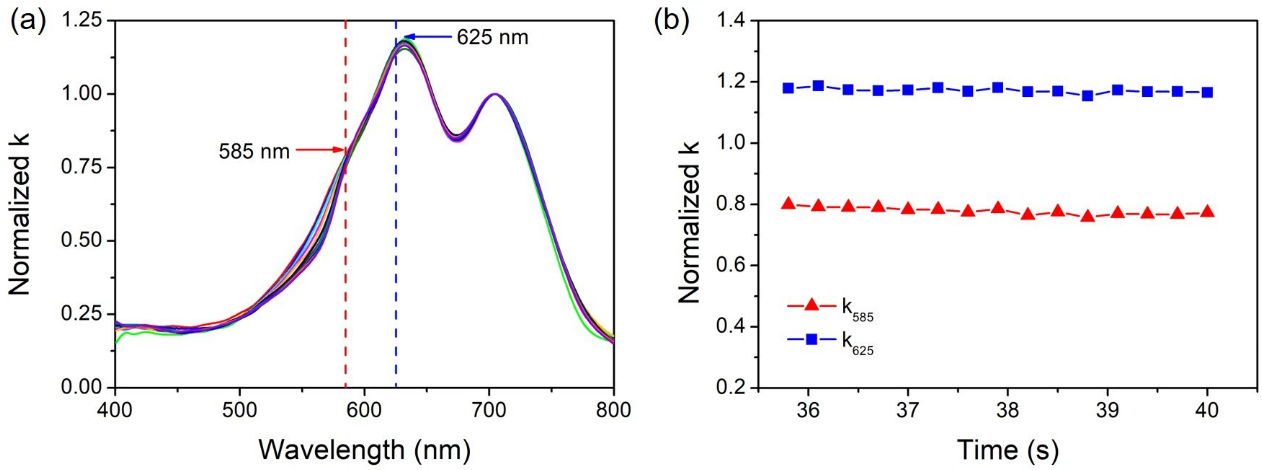


Figure S3. (a) Normalization of the extinction coefficient spectra in stage IV based on the peak of ITIC at 705nm to highlight the relative changes of PBDB-T vibronic peaks at 585 and 625 nm (whose positions are labelled by the blue and red dashed lines). (b) The intensities of extinction coefficients of PBDB-T vibronic peaks (at 585 and 625 nm) vs time extracted from Figure S3a.

5. The extinction coefficient evolution of neat PBDB-T and ITIC.

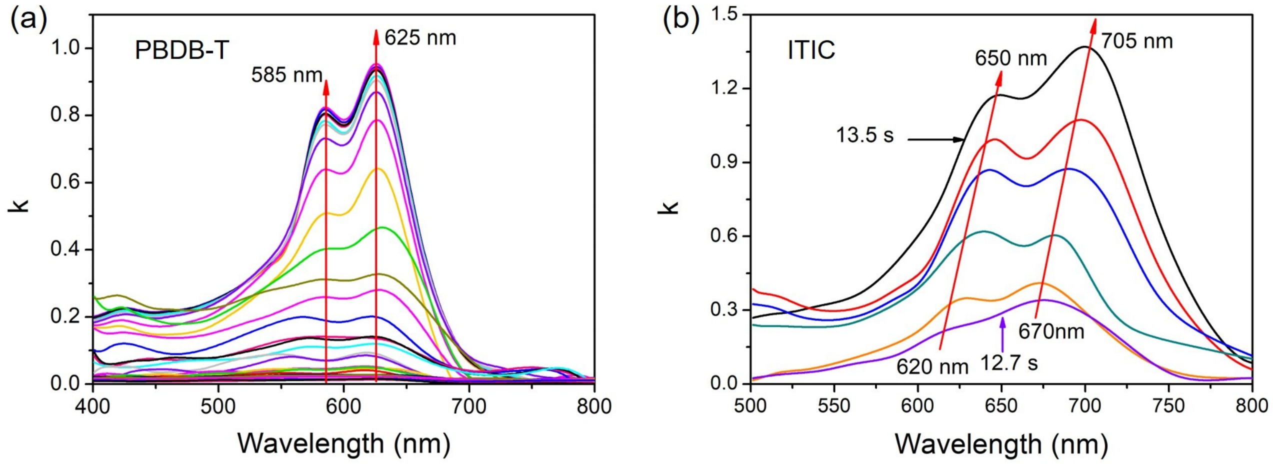


Figure S4. (a) The evolution of k spectra of neat PBDB-T during the film drying process, the vibronic peaks at 585 and 625 nm show no distinct red-shift throughout the process. (b) Part of the k spectra of neat ITIC, the vibronic peaks exhibit clear red-shifts. Note that the onset of red-shift (12.7 s) is consistent with the onset point of increased k rate in neat ITIC drying process.

References

[S1] M. H. Hutchinson, J. R. Dorgan, D. M. Knauss and S. B. Hait, *J. Polym. Environ.*, 2006, **14**, 119-124.

[S2] M. T. Othman, J. A. Lubguban, A. A. Lubguban, S. Gangopadhyay, R. D. Miller, W. Volksen, H. C. Kim, *J. Appl. Phys.*, 2006, **99**, 083503.