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A data-driven identification of morphological features influencing the fill factor and efficiency of organic photovoltaic devices

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ABSTRACT

The performance of organic solar cells is strongly dependent on the morphology of the bulk heterojunction active layer. There has been intense efforts to identify and quantify morphological traits that correlate with various stages of the photo physics. While it is generally accepted that donor domain size affects exciton dissociation efficiency and connectivity affects charge collection, identifying morphology trait(s) that correlate with fill factor and total efficiency have remained elusive. In this work, we utilize correlation analysis on a large set of two dimensional bulk heterojunction morphologies to identify traits that are correlated with fill factor and efficiency. A large dataset of bulk heterojunction morphologies using a phase-field model of phase separation was first created. A comprehensive suite of morphology descriptors were evaluated for each of these morphologies using a recently developed graph based approach. Following this, a morphology aware excitonic-drift-diffusion based device model was used to compute current-voltage curves, fill factors, efficiencies as well as spatial distributions of exciton generation, dissociation, and charge collection for each of the morphologies. We find that (for a given material system with a specified HOMO-LUMO gap, and assuming perfect contact with electrodes) device efficiency primarily depends on the short circuit current, and has almost no dependence on the fill factor. Interestingly, we find that the fill factor is largely insensitive to many of the investigated descriptors. It is only weakly dependent on the contact area mismatch – the difference between the fraction of anode in direct contact with donor and the fraction of cathode in direct contact with acceptor. The fill factor is maximized when this quantity is nearly balanced. Since morphologies with a higher fraction of the electrodes in contact with the desirable material show higher short circuit current, we conclude that designing morphologies for a high short circuit current will necessarily lead to reasonably high fill factors.

1. Introduction

Organic solar cells offer an attractive alternative to inorganics due to their potential low cost, solution processability, light weight, and flexibility. The efficiency of organic photovoltaic cells is now more than 10% [1], thus making them commercially viable, at least for specific niche applications and markets or in conjunction with other devices. It is now understood that the morphology of the active layer has a substantial impact on the photovoltaic performance [2–5]. Several experimental studies have explored the impact of varying morphological features on the photovoltaic performance, specifically the current-voltage (J-V) curve [6–8]. Morphology, in each of these cases, is varied by changing the processing protocol – by using different solvents [9], solvent additives [10–12], solvent annealing [13–16], and thermal annealing [3,17–19]. By changing the degree of crystallinity, domain sizes of the donor and acceptor phases, as well as the amount of donor-acceptor interface, researchers were able to identify morphological features influencing exciton dissociation and charge collection [3,12,20,21]. This has enabled subsequent optimization and control of processing parameters that result in desired morphological features thus producing higher performance active layers and devices [4,7].

In spite of these advances, identification of morphological features that are correlated with the fill factor and total efficiency has remained challenging [2,22]. This is challenging due to several factors, including (a) the possibly non-linear and inter-dependent

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impact of multiple morphological features on these quantities, (b) the dependence on morphological features that are potentially difficult to characterize or measure, and (c) the difficulty in experimentally exploring these process-structure-property relationships, especially given a large set of possible processing conditions. Motivated by these challenges, we deploy a computational approach that utilizes correlation analysis – on a suite of morphological features extracted from a large dataset of representative two-dimensional bulk heterojunction morphologies, along with the simulated current-voltage characteristics of this set of representative morphologies – to identify morphological features that show promising correlation with the fill factor and total efficiency. We explore combinations of various morphological descriptors that have been shown to have some correlation with various measure of performance in 3D simulations [23,24] as well as in experiments [2,25].

This work integrates and builds upon three distinct aspects of research threads pertaining to organic photovoltaics: (a) modeling the evolution of bulk heterojunction morphologies during processing and annealing conditions [26–28], which is used to create a large dataset of representative morphologies; (b) graph-based analysis of morphologies [26,29–32], which is used to extract a suite of morphology descriptors that describe each morphology; and (c) morphology-aware device models based on solving the excitonic drift-diffusion equations [33–35], which is used to generate the current–voltage curves. By using high performance computing resources and integrating these three aspects, we are able to construct a dataset of morphologies, a set of descriptors for each morphology, and the device characteristics of each morphology. Simple data analysis is then used to identify specific morphological features with promising correlations with short circuit current, fill factor and total efficiency.

2. Methods

2.1. Morphology dataset creation

We used our in-house, validated simulation software that models morphology evolution to create a large dataset of representative morphologies. This in-house software is a finite element framework to model multi-physics (evaporation, substrate, fluid shear) driven morphology evolution in multi-component systems that describe the active layer in organic solar cells. This framework generates snapshots of the morphology by modeling the evolution of the morphology under the effect of processing conditions. We used this framework to simulate the evolution of a binary system undergoing thermal annealing in 2D. The binary system consists of an electron donor material and an electron acceptor material. The system undergoes phase separation in response to thermal annealing. The free energy for this system is described by the Flory-Huggins free energy system [27,28]. We consider a computational domain of size 800 nm $\times$ 200 nm. A total of 2000 unique morphologies exhibiting a variety of domain sizes and interfacial areas were selected to populate the dataset. Fig. 1 illustrates several representative morphologies that are produced as a result of these simulations.

2.2. Extracting morphology traits

We utilize a recently developed framework that can extract a comprehensive suite of morphological descriptors. It is based on a graph-based framework to efficiently construct a broad suite of physically meaningful descriptors. These descriptors are classified according to the physical sub-processes of exciton generation, exciton diffusion, charge separation and charge transport. This approach is motivated by the equivalence between a discretized morphology and a labeled, weighted, undirected graph. A detailed discussion of this methodology is provided in Wodo et al. [30]. We extract and annotate each morphology by a large suite of descriptors. Particularly promising descriptors were: (i) fraction of domain that can absorb incident radiation weighted by the exciton generation profile (to characterize absorption), (ii) (Gaussian weighted) average distance from any donor region to the donor-acceptor interface (to characterize dissociation), and (iii) contact area of preferential material with respective electrode (to characterize charge transport). Earlier work has shown that the product of these absorption, dissociation, and charge transport descriptors can be used as a predictor for the short circuit current [29]. We chose around 100 morphologies with a relatively high short circuit current predictor to eliminate very poor morphologies (for instance with several islands). We then calculated the current-voltage curves for these 100 morphologies. Other morphological features we extracted include: donor/acceptor interface area, fraction of interface pixels with complementary paths to both electrodes, the directionality of the donor/acceptor interfaces, fraction of each electrode covered with desirable material, path lengths from interface to electrodes, and finally the tortuosity of the carrier pathways to electrodes.

2.3. Computing device characteristics

We utilize our in-house module that simulates the device physics of a given morphology to compute the current-voltage operation characteristics [34]. This in-house software is based on a finite

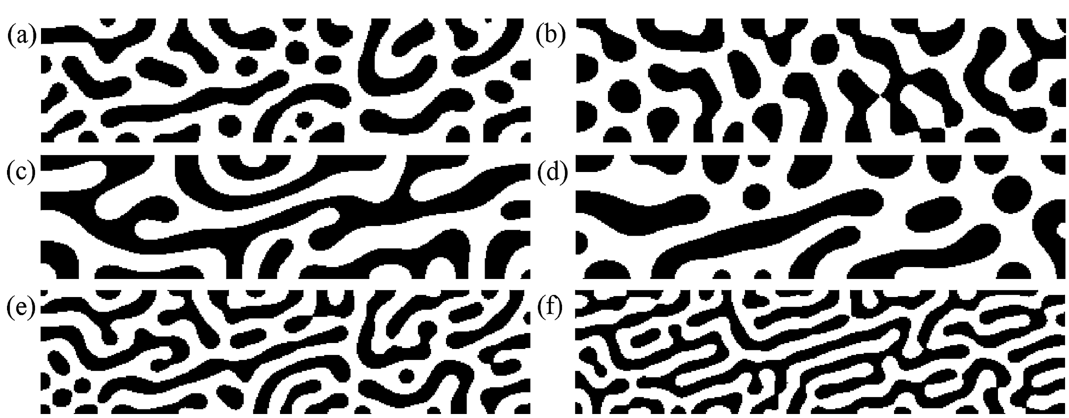

Fig. 1. A representative set of morphologies with a wide range of short circuit current densities (a) 6.7, (b) 7.2, (c) 9.5, (d) 10.3, (e) 12.3, and (f) 16.6 A/m^2, as calculated by the drift-diffusion model.