Competition between recombination and extraction of free charges determines the fill-factor of organic solar cells

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ABSTRACT

Of the parameters that characterize a solar cell, the fill-factor (*FF*) is the least well understood, making targeted improvement difficult. Here, we show that the *FF* of bulk heterojunction solar cells is determined by the competition between charge extraction and recombination. Moreover, we empirically demonstrate the precise relationship between this competition and the *FF* for a wide variety of donor/acceptor combinations. Our finding is supported by experimental measurements on 15 different donor:acceptor combinations as well as by drift-diffusion simulations of organic solar cells in which charge carrier mobilities, recombination rate, light intensity, energy levels, and active layer thickness were all varied over wide ranges to reproduce typical experimental conditions. The results apply to polymer/fullerene and polymer/polymer combinations of different thicknesses, composition, temperatures, and light intensities. For the whole range of 0.26-0.74, all *FF*s follow from the competition between extraction and recombination.

We measure charge carrier mobilities and bimolecular recombination rates using a combination of steady-state and transient extraction techniques. These quantities are used to estimate the extraction and recombination times. The ratio between the extraction and recombination times, which we call theta, indicates whether the majority of charge carriers recombine or can be extracted from the solar cell. If all the *FF*s of the solar cells studied are plotted versus theta, the data collapse onto one universal curve. This shows that the main determinant of the *FF* is the balance between bimolecular recombination and charge extraction.

To explain the observed trend of *FF* versus theta, we perform drift-diffusion simulations of organic solar cells. Charge carrier mobilities, recombination rate, light intensity, energy levels, and active layer thickness were all varied over wide ranges to reproduce the experimental conditions. Again, when the resulting *FF* are plotted versus theta a universal curve is obtained that is in quantitative agreement with the experimental data.

The results presented here provide new insights into the physical phenomena governing the fill-factor of organic solar cells and help explain why the *FF*s change significantly with materials properties, light intensity and thickness. The relationship between *FF* and theta shown by this work offers an approach for targeted improvements of *FF*. In particular, this relationship can be used to rationalise the effect on *FF* of simultaneously changing multiple parameters. Additionally, we indicate in which way recombination and transport properties of a blend should be modified for a device with given thickness, generation rate and internal voltage in order to optimize its *FF*.