

## Relating nanoscale structure to optoelectronic functionality in multiphase donor–acceptor nanoparticles for printed electronics applications

**Mohammed F. Al-Mudhaffer**, Department of Physics, College of Education for Pure Sciences, University of Basrah, Basrah, Iraq; Centre for Organic Electronics, University of Newcastle, Callaghan, NSW 2308, Australia

**Natalie P. Holmes, Pankaj Kumar, Matthew G. Barr,** and **Sophie Cottam**, Centre for Organic Electronics, University of Newcastle, Callaghan, NSW 2308, Australia

**Rafael Crovador**, Centre for Organic Electronics, University of Newcastle, Callaghan, NSW 2308, Australia; School of Biomedical Sciences and Pharmacy, University of Newcastle, Callaghan, NSW 2308, Australia

**Timothy W. Jones**, CSIRO Energy Centre, Mayfield West, NSW 2304, Australia

**Rebecca Lim**, School of Biomedical Sciences and Pharmacy, University of Newcastle, Callaghan, NSW 2308, Australia

**Xiaojing Zhou, John Holdsworth, Warwick J. Belcher,** and **Paul C. Dastoor**, Centre for Organic Electronics, University of Newcastle, Callaghan, NSW 2308, Australia

**Matthew J. Griffith** , Centre for Organic Electronics, University of Newcastle, Callaghan, NSW 2308, Australia; School of Aerospace, Mechanical and Mechatronic Engineering, University of Sydney, Sydney, NSW 2006, Australia

Address all correspondence to Matthew J. Griffith at [matthew.griffith@sydney.edu.au](mailto:matthew.griffith@sydney.edu.au)

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### Abstract

This work investigated the photophysical pathways for light absorption, charge generation, and charge separation in donor–acceptor nanoparticle blends of poly(3-hexylthiophene) and indene-C<sub>60</sub>-bisadduct. Optical modeling combined with steady-state and time-resolved optoelectronic characterization revealed that the nanoparticle blends experience a photocurrent limited to 60% of a bulk solution mixture. This discrepancy resulted from imperfect free charge generation inside the nanoparticles. High-resolution transmission electron microscopy and chemically resolved X-ray mapping showed that enhanced miscibility of materials did improve the donor–acceptor blending at the center of the nanoparticles; however, a residual shell of almost pure donor still restricted energy generation from these nanoparticles.

### Introduction

Organic photovoltaic (OPV) devices have been the focus of intense research for the past two decades. The major benefit of this technology is the potential to deposit the photoactive materials from solution using cheap and scalable printing techniques onto large flexible substrates, thus providing substantial reductions in the manufacturing costs in comparison to other photovoltaic technologies.<sup>[1]</sup> While improvements in the solar-to-electrical power conversion performance in OPV devices have attracted significant attention, progress toward the frequently stated advantage of solution-processable materials that can be dissolved in eco-friendly solvents and scalable printing or coating technologies is commonly overlooked.<sup>[2]</sup> In recent years, an increasing amount of attention has turned toward the printable fabrication of OPV devices as the performance of materials reaches values at which they would be economic at large manufacturing scales and new materials science innovations are developed that allow translation of electroactive materials to large-scale printing equipment.<sup>[3]</sup>

A firmly established criterion for achieving high performance in OPV devices is precise control of the material or device nanostructure in order to achieve the optimal electronic functionality.<sup>[4–6]</sup> The internal nanostructure of OPV materials,

often referred to as the material morphology, is fundamentally complex and is typically achieved with carefully controlled laboratory fabrication and processing techniques.<sup>[7]</sup> However, translating these processes to large area printing whilst maintaining the same level of control over the material nanostructure remains a significant challenge due to the vastly reduced fabrication levers available in the printing process at scale.<sup>[8]</sup> The discrepancy between the need for precisely controlled nanostructure in OPV devices and the difficulty of controlling nanostructure in large-scale printing fabrication is an area that must be addressed by the urgent development of new materials science innovations.<sup>[1]</sup>

The photoactive layer of OPV devices is created by intermixing donor and acceptor semiconductors on the nanoscale, such that all photogenerated excitons are always within a short distance of an interface, where they can be split into free electrons and holes.<sup>[9]</sup> Once created, these free charges typically have poor mobility in organic semiconductors, where the transport occurs through hopping between strongly localized states as opposed to fast transport through a crystalline band with a high density of states.<sup>[10]</sup> This problem is typically addressed by modulating the semiconductor crystallinity through established laboratory-scale solvent and thermal