

Importance of the Donor: Fullerene Intermolecular Arrangement for High-Efficiency Organic Photovoltaics

Submitted by Cl ment Cabanetos on Mon, 06/01/2015 - 14:53

Titre	Importance of the Donor: Fullerene Intermolecular Arrangement for High-Efficiency Organic Photovoltaics
Type de publication	Article de revue
Auteur	Graham, Kenneth R [1], Cabanetos, Cl�ment [2], Jahnke, Justin P [3], Idso, Matthew N [4], Labban, Abdulrahman El [5], Ndjawa, Guy ONGongan [6], Heumueller, Thomas [7], Vandewal, Koen [8], Salleo, Alberto [9], Chmelka, Bradley F [10], Amassian, Aram [11], Beaujuge, Pierre M [12], McGehee, Michael D [13]
Pays	Etats-Unis
Editeur	American Chemical Society
Ville	Washington
Type	Article scientifique dans une revue � comit� de lecture
Ann�e	2014
Langue	Anglais
Date	09/07/2014
Num�ro	27
Pagination	9608-9618
Volume	136
Titre de la revue	Journal of the American Chemical Society
ISSN	1520-5126

Résumé en
anglais

The performance of organic photovoltaic (OPV) material systems are hypothesized to depend strongly on the intermolecular arrangements at the donor:fullerene interfaces. A review of some of the most efficient polymers utilized in polymer:fullerene PV devices, combined with an analysis of reported polymer donor materials wherein the same conjugated backbone was used with varying alkyl substituents, supports this hypothesis. Specifically, the literature shows that higher-performing donor-acceptor type polymers generally have acceptor moieties that are sterically accessible for interactions with the fullerene derivative, whereas the corresponding donor moieties tend to have branched alkyl substituents that sterically hinder interactions with the fullerene. To further explore the idea that the most beneficial polymer:fullerene arrangement involves the fullerene docking with the acceptor moiety, a family of benzo[1,2-b:4,5-b']dithiophene-thieno[3,4-c]pyrrole-4,6-dione polymers (PBDTTPD derivatives) was synthesized and tested in a variety of PV device types with vastly different aggregation states of the polymer. In agreement with our hypothesis, the PBDTTPD derivative with a more sterically accessible acceptor moiety and a more sterically hindered donor moiety shows the highest performance in bulk-heterojunction, bilayer, and low-polymer concentration PV devices where fullerene derivatives serve as the electron-accepting materials. Furthermore, external quantum efficiency measurements of the charge-transfer state and solid-state two-dimensional (2D) $^{13}\text{C}\{^1\text{H}\}$ heteronuclear correlation (HETCOR) NMR analyses support that a specific polymer:fullerene arrangement is present for the highest performing PBDTTPD derivative, in which the fullerene is in closer proximity to the acceptor moiety of the polymer. This work demonstrates that the polymer:fullerene arrangement and resulting intermolecular interactions may be key factors in determining the performance of OPV material systems.

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DOI [10.1021/ja502985g](https://doi.org/10.1021/ja502985g) [15]

Lien vers le document <http://dx.doi.org/10.1021/ja502985g> [15]

Titre abrégé J. Am. Chem. Soc.

Liens

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- [15] <http://dx.doi.org/10.1021/ja502985g>

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