Experimental report

13/09/2022

Proposal: **7-04-168** **Council:** 10/2018

Title: Structural dynamics of bulk heterojunction with non-fullerene acceptors for organic photovoltaics

Research area: Materials

This proposal is a resubmission of 7-04-160

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Samples: $C_{72}H_{88}N_6O_2S_8$
$(C_{10}H_{14}S)_n$
$C_{76}H_{92}N_6O_2S_6$
$C_{72}D_{88}N_6O_2S_8$
$C_{76}D_{92}N_6O_2S_6$

Instrument	Requested days	Allocated days	From	To
IN6-SHARP	6	2	12/09/2019	14/09/2019
IN5	5	0		

Abstract:

In the recent years, the focus in the field of organic photovoltaic has moved from fullerene acceptors that are costly and unstable to nonfullerene acceptors (NFAs). We propose to study the dynamical processes of blends of a conjugated polymer poly(3-hexylthiophene) and two NFAs, IDTBR and IDFBR. They have been reported to give high efficiencies in ternary blends with P3HT. In this proposal, we focus on the 3 blends P3HT:IDTBR, P3HT:IDFBR and IDTBR:IDFBR with the longer aim to study the ternary system.

CHEMISTRY OF MATERIALS

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Structural Dynamics of Polymer:Non-Fullerene Organic Solar Cell Blends: A Neutron Spectroscopy Perspective

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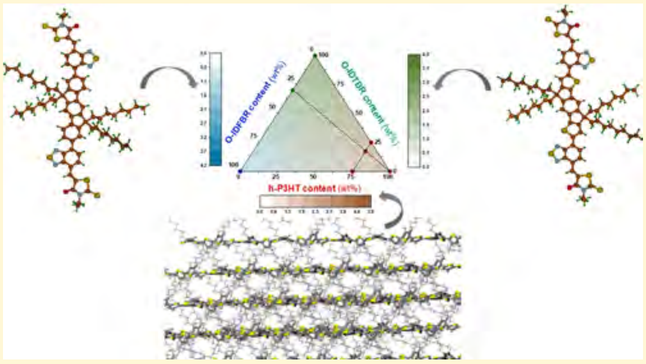
Cite This: *Chem. Mater.* 2022, 34, 7937−7946 Read Online



ABSTRACT: Organic solar cells (OSCs) based on ADA-type (acceptor−donor−acceptor) non-fullerene acceptors (NFAs) exhibit improved power conversion efficiency (PCE) compared to the conventional fullerene-based analogues. The optoelectronic properties of the OSC active layer are correlated to the underlying structural dynamics of the active layer blend and therefore influence the device performance. Using synergistically different neutron spectroscopy techniques, we studied the dynamics of binary and ternary blends made of the non-fullerene acceptors O-IDTBR and O-IDFBR and the regioregular donor polymer poly(3-hexylthiophene) (P3HT). Considering key factors like structural relaxation, miscibility, and morphology, within a chosen temperature range, we probed the dynamical responses of the neat phases and the blends using time-of flight quasi-elastic neutron scattering and neutron spin echo measurements, complemented by neutron vibrational spectroscopy. We cover the femtosecond to nanosecond time window directly relevant to the operating active layer molecular processes. Blends of protonated O-IDTBR and O-IDFBR with either deuterated or protonated P3HT were considered for a contrast variation purpose. Our study confirms the observation of the hypomiscibilty of O-IDTBR in the binary P3HT:O-IDTBR compared to O-IDFBR in P3HT:O-IDFBR and points toward a molecular alloying character of the NFA blend O-IDTBR:O-IDFBR, not observed in the ternary P3HT:O-IDTBR:O-IDFBR. A main outcome of this work is the evidenced similar dynamical response of the donor and acceptor components in both the binary and ternary blends, within the accessible experimental picosecond−nanosecond time window and up to 400 K. This finding is in contrast with our previous neutron spectroscopy and molecular dynamics studies of the fullerene-based blend P3HT:PCBM (PCBM: phenyl-C61-butyric acid methyl ester), where we highlighted distinct behaviors of P3HT and PCBM in the blend in terms of the vitrification/frustration of P3HT upon blending and the plasticization of PCBM by P3HT. Alike P3HT vitrification is not presently observed, which we ascribe to the resemblance of the chemical structures of O-IDTBR/O-IDFBR and P3HT. The absence or the weak vitrification evidenced here is in line with recent reports and is likely related to the improved PCE exhibited by the ADA-type NFA-based OSCs.

■ INTRODUCTION

The active layer of polymeric organic solar cells (OSCs) is composed of an electron-donating polymer and (an) electron-accepting small-molecule semiconductor(s). The electron-accepting organic semiconductors, which have been developed to date, can be broadly separated into two classes: (i) fullerene acceptors and (ii) non-fullerene acceptors (NFAs). Although NFAs were developed early on for vacuum-deposited OSCs, fullerene acceptors have been dominating the field since the introduction of the first solution-processed bulk heterojunction due to their compatibility with a large range of donor materials and relative ease of processing.[1] Early NFAs were suffering from a strong tendency to aggregate due to strong $\pi-\pi$ interactions stemming partially from their 2D planar shape. Thus, large domains and aggregates of acceptors were forming in the active layer leading to poor efficiencies in comparison with fullerene acceptors. Fullerene acceptors are, however, poorly harvesting the solar energy. Recently, ADA-type (acceptor−donor−acceptor) NFAs started to compete with fullerene acceptors.[2] ADA-type NFAs exhibit three structural elements: the core, the end units, and the side chains enabling controlling the energy levels, optical properties, solubility, crystallinity, and electron mobility. In particular, the excessive aggregation observed in earlier NFAs could be controlled by using twisted cores leading to “3D-like” molecules and by engineering bulky ”out-of-plane” side chains. The acceptor groups (end units) can be chosen to ensure good $\pi-\pi$ stacking. Recent efforts have made it possible to synthesize

Received: June 8, 2022
Revised: August 10, 2022
Published: August 18, 2022

https://doi.org/10.1021/acs.chemmater.2c01705