



Solution stability of active materials for organic photovoltaics

Junliang Yang^{a,b,*}, Noel Clark^c, Mengqiu Long^{a,b}, Jian Xiong^{a,b}, David J. Jones^d,
Bingchu Yang^{a,b}, Conghua Zhou^{a,b}

^a Institute of Super-microstructure and Ultrafast Process in Advanced Materials, School of Physics and Electronics, Central South University, Changsha, Hunan 410083, China

^b Hunan Key Laboratory for Super-microstructure and Ultrafast Process, School of Physics and Electronics, Central South University, Changsha, Hunan 410083, China

^c CSIRO Materials Science and Engineering, Private Bag 10, Clayton South, VIC 3169, Australia

^d Bio21 Institute, University of Melbourne, 30 Flemington Road, Parkville, VIC 3010, Australia

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Abstract

The solution stability of the active materials poly(3-hexylthiophene) (P3HT), [6,6]-phenyl C61 butyric acid methyl ester (PCBM), and P3HT:PCBM blends were studied by a combination of organic photovoltaic (OPV) device performance, ultraviolet–visible (*uv–vis*) spectroscopy, and simulation calculations. OPV devices based on the structure ITO/PEDOT:PSS/Active layer (P3HT:PCBM)/Al showed that pure P3HT, pure PCBM, and P3HT:PCBM blend solutions could be stable for as long as one month when stored in either a glove-box or in air. Especially, P3HT:PCBM solution blends are much more stable than P3HT or PCBM solutions stored separately, in which the former could be stored stably over two months. The addition of PCBM to P3HT solution forms P3HT:PCBM dimers, avoiding the formation of charge transfer complexes. The calculation results showed that the electronic wave-function of highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) are isolated in P3HT:PCBM dimers. The research provides a new understanding on the stability of P3HT and P3HT:PCBM solutions, and has great practical application in the fabrication of large-area OPV modules by printing or coating techniques.

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1. Introduction

Organic photovoltaics (OPVs) are continuing to attract considerable attention because of their potential as low-cost and light-weight sources of renewable energy. Power conversion efficiency (*PCE*) has increased rapidly over the last decade, potentially reaching as high as 15%

(Green et al., 2014; He et al., 2012; Dennler et al., 2008; Siddiki et al., 2010). Roll-to-roll printing or coating techniques are preferred for the fabrication of large-area, solution-processable OPV modules with high output and low cost, and so inkjet printing, screen printing, slot-die coating, spray coating, gravure printing, etc., have become hot topics (Krebs, 2009; Tobjörk and Österbacka, 2011; Krebs et al., 2010; Yang et al., 2013; Voigt et al., 2012; Hoth et al., 2007). In addition to device performance and the application process, stability is another factor relevant to the successful commercialization of OPVs. Yu et al. first reported preliminary data on the stability of OPV devices

* Corresponding author at: Institute of Super-microstructure and Ultrafast Process in Advanced Materials, School of Physics and Electronics, Central South University, Changsha, Hunan 410083, China. Tel.: +86 731 88660256; fax: +86 731 88877805.

E-mail address: junliang.yang@csu.edu.cn (J. Yang).

kept in the dark in a glovebox, showing the importance of degradation (Yu et al., 1994). More research was conducted, focused on device stability, degradation mechanisms and device encapsulation (Cumpston and Jensen, 1998; Katz et al., 2001; Schuller et al., 2004; Dennler et al., 2005; Krebs and Norrman, 2007; Jørgensen et al., 2008; Peters et al., 2011; Jørgensen et al., 2012). Degradation has two main aspects, *i.e.*, chemical degradation and physical degradation. The chemical degradation of OPV devices is related to chemical reactions resulting from the diffusion of oxygen and water into the electrode material and into the active polymer layer, as well as interface problems, especially photochemistry and photo-oxidation; while physical degradation is related to changes in microstructure, such as morphology, phase separation and the bending of flexible OPVs, leading to a reduction of OPV performance (Jørgensen et al., 2008, 2012). The degradation mechanisms and processes in OPV devices are very complicated because there are many factors acting. It is useful to study the degradation of each component of OPVs separately, for example, the degradation of the active layer material or the degradation of the electrode. However, almost all degradation studies are focused on complete OPV devices, in which both chemical degradation and physical degradation occur, including at the electrode, in the active layer, the interface, etc.

The study of active material solution stability has both practical application and theoretical utility to the development of OPVs. It not only helps us understand the stability of OPV devices, but it also provides solution stability information that is relevant to the production of large-area OPV modules fabricated by printing or coating techniques. However, no systematic studies currently exist. In present paper, we provide a detailed study of the solution stability of the active materials poly(3-hexylthiophene) (P3HT), [6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM), and P3HT:PCBM blends, with respect to the performance of organic photovoltaic (OPV) devices, ultraviolet–visible (*uv–vis*) spectroscopy, and simulation calculations. The research results suggest that the solutions have different stabilities. P3HT:PCBM blend solutions are much more stable than pure P3HT or PCBM solutions stored separately, either in a glovebox or in air, because P3HT and PCBM can stabilize each other in the blend solution. In particular, we show how the degradation of the active materials influences the performance of OPV devices when other factors are excluded. More importantly, our study of the stability of active layer material solutions provides information relevant to the commercial fabrication of large-area OPV modules when printing or coating techniques are used in air.

2. Experimental

2.1. Materials

Poly(3-hexylthiophene) (P3HT, EE99002, Merck) and two types of [6,6]-phenyl C₆₁ butyric acid methyl ester,

comprising technical grade (TG-PCBM, 99%, Solenne BV) and research grade (RG-PCBM, 99.5%, Nano-C Inc.), were combined to form the active layer in OPV devices. ITO-coated glass with a sheet resistance of 5 Ω/□ was purchased from Lumtek. Aluminium pellets (99.999%) were purchased from KJ Lesker. An aqueous dispersion of poly-3,4-ethylenedioxy-thiophene:poly(styrene sulphonic acid) (PEDOT:PSS, Clevios P VP Al 4083, Heraeus) was the hole transport layer and was used as received.

2.2. Solution fabrication

In order to obtain stability information for pure P3HT, pure PCBM and P3HT:PCBM blend solutions, a 3-month experiment was designed. The groups of solutions shown below were made on the same date using chlorobenzene (CB) as the solvent: (i) 10 ml P3HT:RG-PCBM blend solution (15 mg/ml + 12 mg/ml) kept in a glovebox, (ii) 10 ml P3HT:TG-PCBM blend solution (15 mg/ml + 12 mg/ml) kept in a glovebox, (iii) 10 ml P3HT (30 mg/ml), 5 ml RG-PCBM (24 mg/ml), and 5 ml TG-PCBM (24 mg/ml) solutions kept in a glovebox, separately, (iv) 10 ml P3HT:RG-PCBM blend solution (15 mg/ml + 12 mg/ml) kept in air, (v) 10 ml P3HT:TG-PCBM blend solution (15 mg/ml + 12 mg/ml) kept in air, (vi) 10 ml P3HT (30 mg/ml), 5 ml RG-PCBM (24 mg/ml), and 5 ml TG-PCBM (24 mg/ml) solutions kept in air, separately. The solutions kept in air were prepared in air, and the solutions kept in a glovebox were prepared in a glovebox. All the solutions were placed in vials with stirrers and the caps sealed, and were then kept under ambient light and temperature either in air or in a glovebox. The temperature and light illumination levels in both cases were comparable.

2.3. Device fabrication

Before making OPV devices, the solutions were heated to 60 °C and stirred for 30 min in a glovebox (for the solutions kept in glovebox) or in air (for the solutions kept in air). If the P3HT and PCBM were stored separately, they were mixed in a ratio of 1:1 for each experiment before the fabrication of the OPV devices. Hence, there were 8 types of devices: (A) P3HT:RG-PCBM blend solution stored in a glovebox, (B) P3HT:TG-PCBM blend solution stored in a glovebox, (C) freshly mixed P3HT and RG-PCBM stored in a glovebox, (D) freshly mixed P3HT and TG-PCBM stored in a glovebox, (E) P3HT:RG-PCBM blend solution stored in air, (F) P3HT:TG-PCBM blend solution stored in air, (G) freshly mixed P3HT and RG-PCBM stored in air, (H) freshly mixed P3HT and TG-PCBM stored in air.

Two devices with 12 cells were fabricated for each solution type and storage time. ITO-coated glass was cleaned by successively sonicating for 10 min each in distilled water, acetone and iso-propanol. The substrates were then exposed to a UV-ozone clean (at RT) for 10 min. The

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