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Impact of environmentally friendly processing on polymer solar cells: Performance, thermal stability and morphological study by imaging techniques



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ABSTRACT

The combination of mass-production compatible coating techniques and environmentally friendly solvents to process bulk heterojunction solar cells represents a key issue to scale up this technology. In this work we demonstrate that using a benchmark polymer HBG-1 blended with PC₆₁BM, the replacement of a common chlorinated processing solvent (orthodichlorobenzene) with a non-chlorinated analogous (*o*-xylene) not only allows the fabrication of blade-coated bulk heterojunction devices with identical photovoltaic performance, but also determines a great enhancement of the resulting thermal stability. Thermal degradation tests were carried out in inert atmosphere, by keeping the solar cells onto a hot plate at 85 °C and monitoring their OPV performance. In parallel, the morphological changes of the active layers induced by thermal stress are investigated by combining two complementary light-based imaging techniques, laser scanning confocal and photocurrent microscopy, which offer the great advantage to simultaneously study on complete devices the blend morphology and the electrical properties, point-by-point, of the active layer even in regions unlikely accessible (e.g. the active area under the top electrode) using other techniques. As a result, we found that solar cells processed from a non-chlorinated based solvent, in comparison to an analogous reference system, exhibit a different evolution of the resulting BHJ morphology during thermal ageing, in perfect agreement with the corresponding photovoltaic responses.

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1. Introduction

With the increasingly urgent need for clean energy production, in the last decades photovoltaic technologies has strongly evolved. In this context, organic solar cells (OSCs) have attracted much interest due to their characteristic of low production costs, low modules weight and color adjustment [1]. One of the most promising class of OSCs are bulk heterojunction (BHJ) ones, in which the active layer is composed of a finely intermixed blend of an electron donor (D) and an electron acceptor (A) material.

The power conversion efficiencies (PCEs) of laboratory-scale BHJ OSCs have been greatly improved surpassing the 10% milestone [2] through the synergic development of novel materials [3], enhanced understanding of polymeric film microstructure [4] and

meticulous device optimization [5], indicating a bright future for innovative commercial applications [6]. Despite the great advantages in terms of PCEs, other fundamental aspects such as processing conditions and lifetime of the resulting devices need to be seriously considered for the upscale of OSCs toward industrialization and practical applications.

Recent works highlight the great effort of several research groups on the processing methods of OSCs, demonstrating for instance, that the use of (laboratory-scale) deposition techniques fully compatible with roll-to-roll mass production [7] and/or the replacement of common chlorinated processing solvents with alternative ones, having reduced costs, toxicity and environmental impact, represent strategic aspects for OSCs to succeed as a technology [8]. Importantly, the optimized materials and processes should also provide devices/modules with operational stability suitable for the technological requirements, however during their normal functioning OSCs are still subjected to significant degradation processes occurring at various rates and regions of the device. Despite water and oxygen diffusion within the device can

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be prevented through a proper encapsulation, parallel degradation mechanisms induced by light exposure and temperature cannot be avoided [9]. In particular, the behavior of BHJ OSCs with thermal degradation is generally correlated to morphological changes of the active layer, which can influence the efficiency of internal mechanisms responsible for the charge generation, diffusion and collection processes [10]. This kind of degradation and thus the resulting BHJ stability are strongly dependent on the initial nanoscale morphology of the active layer, which is mainly dominated not only by the structure/nature of the components but also by the relative processing conditions. Indeed, several key factors including solubility, miscibility, solvent evaporation rate, supersaturation thermodynamics and precipitation/segregation kinetics, all aspects responsible for the self-organization of the BHJ components, are strictly related to the chemico-physical properties of the processing solvent [11]. In addition it is worth mentioning that beside the intrinsic properties of the generated BHJ blends, the resulting surface interactions with the adjacent buffer layers and/or electrodes can further contribute to the stabilization of the BHJ morphology [12].

In this context, the impact of relatively cheap and environmentally friendly solvents on the morphological and electrical properties of corresponding OSCs has been recently investigated for different polymer:fullerene blends [13]. However, additional effects/benefits of this approach, such as the impact on the thermal stability of the corresponding OSCs, has not been explored yet.

Here we demonstrate that, using a benchmark polymer (HBG-1) as donor material, the replacement of a common chlorinated processing solvent (orthodichlorobenzene, ODCB) with a non-chlorinated analogous (*o*-xylene) not only allows the fabrication of blade-coated BHJ solar cells with identical performance, but also determines a great enhancement of their morphological stability against the thermal stress. To investigate the local morphological features/changes of complete BHJ solar cells before and after thermal ageing, we exploited the potential and versatility of two complementary light-based imaging techniques implemented on the same instrumental setup: LSCM (Laser Scanning Confocal Microscopy) and LSPM (Laser Scanning Photocurrent Microscopy). Their non-invasiveness allows to reliably monitor *in situ*, at different degradation stages, both the evolution of the BHJ morphology and the electrical properties of the active layer in the same working device. Note that the defects and phase segregated domains which usually form and evolve during the thermal stress in the BHJ layer (often in the hundreds of nanometers or microns length scale [14]) perfectly match with the resolution of these techniques.

Unlike the most commonly used methods such as scanning probe, X-ray and electron microscopy, which offer a great potential in terms of resolution [15], the real advantage of light-based techniques is represented not only by the possibility to study complete devices but also to explore crucial regions of the active layer unlikely accessible (without damaging the system) [16], such as the active area under the top electrode. In this way, the interactions occurring at the BHJ/electrode interface and their effect on the evolution of the morphology of the active layer during degradation (confinement effect [12]) can be taken into account. As a result, both for ODCB and *o*-xylene processed devices we found, after thermal ageing, different morphological rearrangements in the areas under and outside the overlapping top electrode, demonstrating not only the key role of the processing solvent and confinement effect on the BHJ morphological evolution, but also a direct and realistic correlation with the corresponding photovoltaic responses.

2. Experimental section

2.1. Materials

Moderate band gap polymer 1 (HBG-1) was provided by Merck and the solution of ZnO nanoparticles (Lab'Ink Jet ZnO) was provided by Genes'Ink. [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM), 1,8-diiodooctane (DIO), MoO₃, Ag and the different solvents were purchased from commercial sources and used without further purification.

2.2. Photovoltaic devices fabrication

Device architecture is glass/ITO/ZnO/active layer/MoO₃/Ag. Patterned ITO-coated glasses ($R_s \sim 10 \Omega \text{ sq}^{-1}$) were cleaned in sequential sonicating baths (for 15 min) in deionized water, acetone and isopropanol. After the final sonication step, substrates were dried with a stream of Ar gas and then placed in an oxygen plasma chamber for 10 min. Next, a thin layer of ZnO ($\sim 30 \text{ nm}$) was deposited by blade-coating (in air) on the top of the glass/ITO substrate and subsequently annealed at 80 °C for 5 min to remove residual traces of solvents. The active layers were prepared from solutions of HBG-P1:PC₆₁BM (1:2 wt/wt) dissolved in orthodichlorobenzene without or with 2.5% v/v of DIO (Device 1 and 2, respectively) or *o*-xylene without or with 2.5% v/v of DIO (Device 3 and 4, respectively) with a total concentration of 30 mg/ml. The active solutions were deposited by blade-coating in air. The films were transferred inside a metal evaporator and kept under high vacuum ($\approx 1 \times 10^{-6}$ Torr) for 15 h, in order to remove the residual amount of solvents and or DIO. To complete the devices, MoO₃ (10 nm) and Ag (90 nm) were subsequently deposited.

2.3. Photovoltaic devices characterization

The current–voltage (*I*–*V*) characteristics of all BHJ devices were recorded by a Keithley 236 source-measure unit under simulated AM1.5G illumination of 100 mW cm⁻² (Abet Technologies Sun 2000 Solar Simulator) inside the glove box. The active area of the devices is 6 mm². The devices were illuminated through a calibrated mask to avoid the parasitic photocurrent arising from the areas outside the electrodes. The thickness of the various films was measured by a profilometer (KLA Tencor, P-6). Atomic Force Microscopy (AFM) images were taken with a Solver Pro (NT-934 MDT) scanning probe microscope in tapping mode. The AFM images were recorded directly on tested devices.

2.4. Thermal ageing conditions

The thermal degradation tests were carried out inside the glove-box, by keeping the devices onto a hotplate at 85 °C for 150 h and measuring, after that time, their *I*–*V* characteristics.

2.5. LSCM and LSPM setup

The LSCM and LSPM images were taken with a Nikon TE2000 optical microscope, equipped with a 60× objective with 0.70 numerical aperture, connected with a Nikon EZ-1 confocal scanning head. They were carried out on both freshly prepared and aged devices to monitor the evolution of the active layer morphology with thermal degradation. All the devices were first encapsulated in the glove box before being measured, to minimize degradation due to water and oxygen ingress. The encapsulated devices were illuminated through the glass/ITO side. The laser excitation wavelength was 488 nm for LSCM and 405 nm for LSPM. To note, the active layer shows a good absorbance (> 0.4) both at 488 nm and 405 nm. In order to obtain images with a higher

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