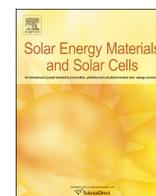




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## 8.7% Power conversion efficiency polymer solar cell realized with non-chlorinated solvents

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## ABSTRACT

The use of environmental friendly solvents for the fabrication of solution processed organic photovoltaics is a key issue to scale up the technology. Nowadays however, toxic and harmful chlorinated solvents are largely used in polymer solar cell laboratory research. In this work we successfully reached high solubility and miscibility of the low band gap polymer Poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]] (PBDTTT-E-F, commonly known as PTB7), blended with [6,6]-Phenyl-C71-butyric acid methyl-ester ([70]PCBM fullerene derivative) in a non-chlorinated solvent (Dimethylbenzenes also known as Xylenes). We studied the solar cells realized depositing blend solutions based on various Xylenes (*ortho*, *para* and an isomeric mixture from technical grade) achieving high power conversion efficiencies up to 8.7%.

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## 1. Introduction

Low band-gap polymer-based Bulk-Heterojunction (BHJ) organic solar cells (OSCs) have gained significant attention in the last few years, since the possibility to reach power conversion efficiencies (PCE) higher than 9% for a single layer device [1], and to overcome the target of 10% in a tandem structure [2].

Such high efficiency values together with the native opportunity to scale up the realization process on large area at relatively low costs [3], push the BHJ-based photovoltaics to the top of the future feasible renewable energy technologies.

There are many applicable printing techniques (screen, spray, gravure and ink-jet printing, blade and slot-die coating) [4–6] that can be implemented on roll-to-roll (R2R) [7,8] mass production systems that make this technology attractive and even if the reproduction of the performance of a laboratory device on large area (module) is still an issue [9], some paper demonstrated feasibility to overcome this obstacle [10,11].

However, the use of toxic and harmful chemicals inside formulation of polymer/fullerene blend printed inks, in particular chlorinated solvents (*chloro*- and *dichloro-benzene*) [12], is

currently the most common choice for high performance devices [1]. The use of these solvents leads to an increase of the overall costs of the large area process because the huge volume quantities involved in the scaling-up are significant and require appropriate design of the production line with suitable safety equipment and specific disposal of hazardous waste [13]. In this context, the challenge is to obtain high efficiencies with the use of inks based on safe and industrial compatible solvents.

In this work, we successfully obtained high performance BHJ inverted solar cells based on a mixture of PTB7 and [70]PCBM (1:1.5), blended at 2.2 wt% in different formulations of Dimethylbenzene. In particular, we compared devices realized with *ortho*-Xylene (*o*-Xy), *para*-Xylene (*p*-Xy), plus a mixture of Xylene isomers (*mix*-Xy), resulting in a chlorine-free processing – except for the small amount (3% v/v) of 1,8-diiodooctane (DIO) additive [14]. We were able to reach an overall high PCE of 8.73%, 7.87% and 8.22%, respectively for *o*-Xy, *p*-Xy and *mix*-Xy. As reference cell in conventional Chlorobenzene (Cb) solution, we obtained a PCE of 7.9%. Furthermore the transmittance of the best performing cell (in the visible + near IR range), resulted increased of the ~21% more than the reference device; thus, it results an enhanced transparency for the final device. This feature would play an important role both for eventual building integration and for the realization of a tandem architecture where the transparency of the first stacked device is a critical issue to achieve high efficiency [15–17].

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## 2. Experimental

### 2.1. Materials

PEIE (Polyethylenimine, 80% Ethoxylated,  $M_w \sim 70,000$  g, Aldrich) was diluted in 2-methoxyethanol (0.2 wt%), as described in literature [15,18,19]. PTB7 and [70]PCBM (99.99%) were purchased from Solarmer and Solenne BV, respectively. *Ortho*-Xylene, *para*-Xylene, molybdenum (VI) oxide ( $\text{MoO}_3$ , 99.98% powder) and silver (Ag, wire  $\geq 99.99\%$ ) were purchased from Sigma Aldrich. The Xylenes isomeric blend mixture is technical grade.

### 2.2. Device fabrication

The devices are realized on ITO glass-covered substrates (Kintex  $\sim 8\Omega/\square$ ), patterned with wet-etching in hydrobromic acid and cleaned in ultrasonic bath with acetone and ethanol (10 min each step). The substrates are covered with PEIE deposited via spin-coating in air under chemical hood at 5000 rpm for 60 s. The PEIE is thermal treated at  $120^\circ\text{C}$  on a hot-plate for 10 min in air [20]. Subsequently, samples are transferred inside a glove box with controlled  $\text{N}_2$  atmosphere to be spin-coated with PTB7:[70]PCBM mixture. The polymer content of PTB7 and [70]PCBM is combined in a ratio of 1:1.5 and dissolved at 2.2% in weight in three different Xylenes: *ortho*-Xylene (1,2-dimethylbenzene), *para*-Xylene (1,4-dimethylbenzene) and a mixture of Xylenes (technical grade), composed of 90% of *ortho*, *meta*, *para*-Xylene (prevalently *ortho*-Xylene), and 10% of a mixture of Ethylbenzene and Toluene where the maximum amount of toluene is 0.5%. Finally the 3%v/v of 1,8-diiodooctane (DIO) is added in the solutions that is then put on stirring overnight at room temperature. As reference, a solution in chlorobenzene is prepared in the same conditions. The active blend is spin-coated at 1000 rpm for 120 s, for a resulting thickness of  $\sim 80$  nm in case of Xylenes films and  $\sim 100$  nm for chlorobenzene one, and subsequently treated in slight vacuum ( $10^{-1}$  mbar) for 20 min to accelerate the drying process and remove residual DIO from thin films [21].

The  $\text{MoO}_3$  hole transporting layer (HTL), and the Ag anode are thermally evaporated at  $10^{-7}$  mbar for a final thickness of  $\sim 5$  nm and  $\sim 100$  nm, respectively. At the end 8 devices of  $0.1\text{ cm}^2$  are defined on each substrate.

### 2.3. Characterization

The thickness of layers was evaluated by a profilometer (Dektak 150). The surface morphology of the thin film in different solvent mixtures was taken with an A.P.E. Research Atomic Force Microscope (AFM). Measurements were performed in *Non-Contact* mode

with a Silicon tip with a radius of 8 nm, mounted on a cantilever (resonance frequency = 325 kHz) with a spring constant of 40 N/m. Scanning transmission electron microscopy (STEM) images have been realized using a FE-SEM platform (Zeiss, Auriga) with an integrated STEM system.

Device electrical ( $J$ - $V$ ) performances were characterized outside glove box with a source-meter (Keithley 2420) under an AM1.5 Class A ABET solar simulator ( $100\text{ mW}/\text{cm}^2$ ); the EQE measures were performed with an IPCE (Incident Photon-to-current Conversion Efficiency) system (IPCE-LS200, Dyers) calibrated with a UV-enhanced Si detector (Thorlabs, 250–1100 nm).

## 3. Results and discussion

We performed a comparison between Chloro- and Dimethylbenzene (Xylene) solution based devices. We considered an inverted structure (Fig. 1a), largely demonstrated to be more reliable and performing [20,21] with respect to the conventional one. This architecture reached the best record efficiency on a single layer with a low band gap polymer [1].

We focused on the active layer film's performance and characteristics: starting from the same concentration in weight of the PTB7:[70]PCBM material content, we carried out four set of samples changing the solvent. For each solvent we chose the thickness of the active layer that led to the best performing device.

The film deposition resulted in all cases regular and pinhole-free all over the substrate before and after vacuum treatment. A typical characteristic of the Xylenes' films is the frame on the border that makes them distinguishable from chlorobenzene one (Fig. 1b). Despite equal spin-coating parameters, the films deposited from Xylene solvents, present an average thickness of  $\sim 80$  nm, instead of  $\sim 100$  nm typical of the Cb one. Differences are evident by looking at the absorbance spectra, where the typical absorption peaks of PTB7 are recognizable in all the films even if they are of lower intensity for Xy films (Fig. 2). The absorbance spectra of Xylene films are completely overlapped due to the similar solvent parameters and thickness of the samples. In addition, the realized samples (Fig. 1b) appear more transparent ( $\sim 62\%$  average transmittance) than the Cb counterpart ( $\sim 55\%$  average transmittance) even if the performance is comparable, even improved.

This result is explainable considering the different viscosity of the two types of solution. In fact, even if Xylenes and Chlorobenzene have similar viscosity ( $\sim 0.80$  cP at  $20^\circ\text{C}$ ) and similar boiling point (*o*-Xy  $144^\circ$ , *p*-Xy  $139^\circ$ , *mix*-Xy  $138^\circ\text{C}$  and Cb  $132^\circ\text{C}$ ), they have different densities (Xy =  $0.88\text{ g}/\text{ml}$ , Cb =  $1.11\text{ g}/\text{ml}$ ). This leads to a different solid content of the solution and therefore to a lower viscosity for the Xylene-based inks.

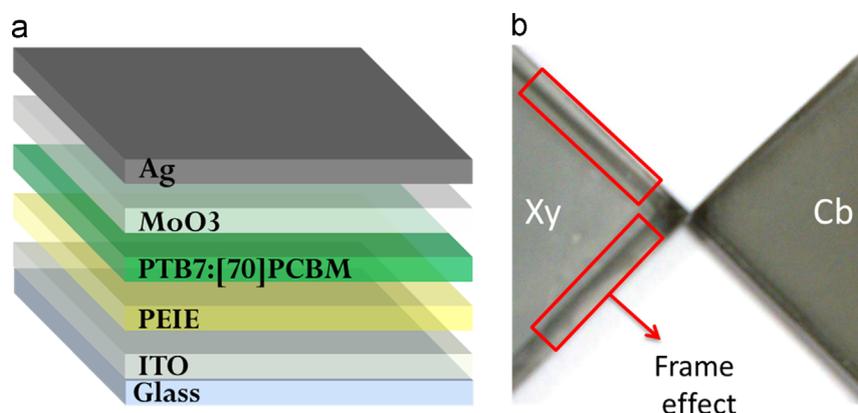


Fig. 1. Stack of the device (a), border effect of Xylene film compared with Chlorobenzene (b).

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