

Predicting thermal stability of organic solar cells through an easy and fast capacitance measurement

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ABSTRACT

Degradation of organic photovoltaic (OPV) devices is currently a topic under intense research as it is one of the main limitations towards the commercialization of this technology. Morphological changes at both active layer and interfaces with the outer contacts are believed to determine main key issues to be overcome. In-line techniques are essential to rule out any effect arising during sample fabrication. Unfortunately, the number of physical techniques able to provide morphological information on complete and operational devices is certainly limited. In this work, we study the thermal degradation of bulk heterojunction (BHJ) solar cells composed by different donor polymers with techniques developed to provide in-situ information on operational devices. Capacitance measurement as a function of temperature monitors the electrical integrity of the active layer and provides the threshold temperature (T_{MAX}) at which the whole device becomes thermally unstable. We found a direct correlation between the threshold temperature T_{MAX} , obtained by capacitance–temperature measurements on complete OPV devices, and the power conversion efficiency decay measured at 85 °C. Devices tend to be thermally stable when the temperature of the thermal stress is below T_{MAX} , while above T_{MAX} evident changes in the active layer or at the active layer/electrode interface are also detected by confocal fluorescence microscopy. The capacitance method gives precious guidelines to predict the thermal stability of BHJ solar cells using an accelerated and easy test.

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1. Introduction

Organic photovoltaic (OPV) devices have to fulfill two fundamental requirements to be competitive in the field of renewable energy sources: (i) power conversion efficiency (PCE) over 10% [1,2] and (ii) lifetime of at least 7–10 years [3]. Recently, laboratory-scale bulk heterojunction (BHJ) solar cells have reached the milestone of 11% [4], through a synergic development of increasingly high performing photoactive materials [5–9], understanding of the morphological film nanostructure [10,11] and device structure optimization [12–18]. Further efficiency improvement is

expected by the use of innovative architectures with improved light absorption such as the use of metasurfaces [19] or layer-by-layer nanoarchitectonics [20]. However, the achievement of high performance has little technological impact if the resulting device lifetime is unsuitable for the technological requirements. For outdoors applications thermal degradation is a key factor that needs to be controlled, as solar panels usually reach temperatures as high as 65–85 °C [21]. Recent studies on several highly efficient OPV devices showed severe efficiency losses even after a short operation time [22]. For this reason, a number of research groups are focused on boosting the lifetime of OPV devices through a deeper study of the degradation mechanisms, attempting to identify, prevent and/or limit them.

Krebs et al. [23] have comprehensively reviewed the most common degradation mechanisms taking place in organic

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photovoltaic. Different physical agents such as humidity, oxygen, UV light and temperature exposure need to be taken into account when studying the degradation of operating OPV devices. Diffusion of water [24] and molecular oxygen [25–27] into the device promotes chemical degradation of interfaces [25] and active material [28]. These effects can be discarded if a device is properly encapsulated. However, degradation pathways due to light soaking and high temperature cannot be eliminated, and, in general, they induce morphology evolution of the active layer [29], interlayer and electrode diffusion [30], and electrode interaction with the organic materials [31]. The behavior of BHJ OPVs after thermal degradation is generally correlated to morphological changes occurring in the active layer that can affect: (i) charge separation process by formation of fullerene aggregates in polymer:fullerene blends, which leads to a PCE loss due to the reduction of the donor:acceptor (D:A) interfacial area [32–34], (ii) charge extraction by a migration of a skin-layer of either polymer [35] or fullerene [36] adhering to the top contact, generating barriers or selective transport regions depending on the device architecture, (iii) transport properties by modification of the polymer packing in the blend [37,38], (iv) recombination by an increase of the number of defect states in the bulk of the active layer [39], and (v) optical properties by generation of a charge transfer complex between donor and acceptor molecules which also acts as an exciton quencher [40]. Several works [33,34] demonstrated that morphological reorganization processes occur only if the solar cell is subjected to temperatures near or above the glass transition temperature (T_g) of the donor polymer. At this temperature devitrification of the blend allows the polymer and the fullerene molecules to rearrange/diffuse in the bulk and at the interface with the electrodes. Typically the T_g of a polymer/blend is measured by Differential Scanning Calorimetry (DSC) [41] but other techniques like ellipsometry have also been employed [38,42]. However, experimental T_g value is not always a well-defined parameter and in many systems a clear transition is not observed. Moreover, this value depends on several aspects such as the thermal history of the sample, the technique/method followed and the thermal rate used for the measurement [43]. For example, recent experiments [44,45] and simulation [46] studies have demonstrated a pronounced thickness-dependent confinement effect on T_g , together with a strong dependence on the solvent and on the substrate used [47]. These effects are totally disregarded if a technique like DSC is employed. On the other hand, ellipsometry can account for this thickness dependence but the presence of the buffer layers and of the electrodes is still not taken into account. In fact, the morphological rearrangement of the BHJ blend which occurs at temperatures above T_g may not be the same in presence or in absence of the top electrode, as demonstrated by several works with the so called “confinement effect” [48]. Taking into account all these considerations, it emerges that it is difficult to predict the thermal behavior of a BHJ solar cell through the bare analysis of the T_g of pristine materials and/or blend films (e.g. measured by DSC or ellipsometry). Indeed, to have a good description of the thermal stability of a solar cell it is crucial to study the properties of the complete device, taking into account the contribution of each layer and each interface on the degradation processes at the same time.

In this work, we study the thermal behavior of different BHJ solar cells using techniques enabling a direct investigation on working devices, such as: (i) current density–voltage characterization to control the evolution of the photovoltaic parameters during the thermal degradation, (ii) capacitance measurements as a function of temperature to monitor the electrical integrity of the active layer, and (iii) fluorescence imaging by Confocal Microscopy to provide visual evidences. Five donor polymers, P(1)-FQ-BDT-4TR, P(2)-PTB7, P(3)-P3HT, P(4)-PTT-MIM, and P(5)-PBnDT-FTAZ

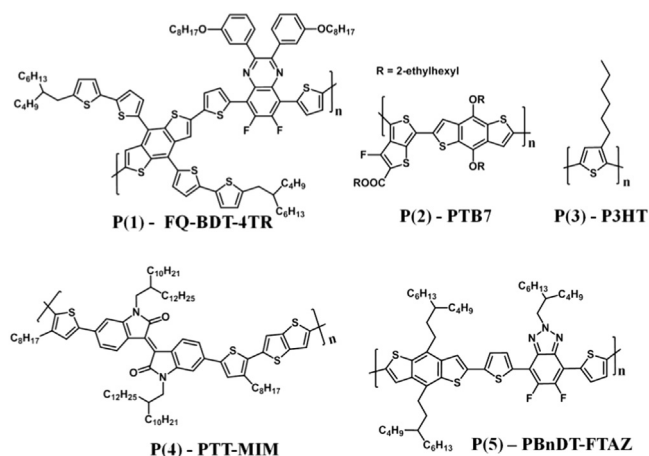


Fig. 1. Chemical structure of the photoactive polymers.

(Fig. 1), which represent a broad range of different types of conjugated polymers, were selected and specific properties of the corresponding BHJ devices were characterized and correlated with the thermal degradation. As a result, we found a direct correlation between the threshold temperature T_{MAX} , obtained by capacitance–temperature measurements on complete OPV devices, and the PCE decay profile measured at 85 °C. In summary, this methodology provides a threshold temperature at which the whole device becomes thermally unstable, thus giving precious guidelines to predict the thermal stability of BHJ solar cells using accelerated tests.

2. Experimental section

For this study we used a standard device architecture: glass/ITO/PEDOT:PSS/Active Layer/ZnO/Ag. The PEDOT:PSS (poly(3,4-ethylenedioxythiophene):poly(4-styrenesulfonate), Clevis P VP A1 4083, H.C Starck) was spun-cast on a pre-cleaned patterned ITO-coated glasses ($R_s \sim 10 \Omega/\square$) to form a thin layer (~ 30 nm) and subsequently annealed at 150 °C for 15 min. Different active layers based on the donor polymers P(1)-FQ-BDT-4TR, P(2)-PTB7, P(3)-P3HT, P(4)-PTT-MIM, and P(5)-PBnDT-FTAZ were prepared. As acceptor materials we used PC₆₁BM ([6,6]-phenyl-C₆₁-butyric acid methyl ester, Solenne BV) or PC₇₁BM ([6,6]-phenyl-C₇₁-butyric acid methyl ester, Solenne BV). The active layers were prepared by spin coating inside the glove-box. The following devices were prepared using recipes reported in literature: P(1):PC₆₁BM (1:1 w/w) [49], P(2):PC₇₁BM (1:1.5 w/w) [12], P(3):PC₆₁BM (1:0.8 w/w) [50] and P(5):PC₆₁BM (1:2 w/w) [51]. On the other hand, P(4):PC₆₁BM (1:1.5 w/w) was deposited from a solution of 1,2-dichlorobenzene and 1,8-Diiodooctane (98:2 v/v) with a total concentration of 30 mg/ml by spinning at 700 rpm for 120 s. Details of the synthesis of P(4) are reported in the [Supporting information](#).

The ZnO (provided by Genes'Ink, Lab'Ink Jet ZnO) was spun cast (inside the glove-box) on the top of the active layer to get a thickness of 50 nm. To complete the device fabrication, Ag (100 nm) was next deposited in high vacuum ($\approx 1 \times 10^{-6}$ Torr) using a thermal evaporator directly connected to the glove-box. The current–voltage (I – V) characteristics of all OPV devices were recorded by a Keithley 236 source-measure unit under simulated AM1.5G illumination of 100 mW/cm² (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

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