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The role of donor polymer and PEDOT:PSS formulation on adhesion processes in inverted organic solar cells



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ABSTRACT

Organic photovoltaic devices (OPVs) are one of the most promising applications of organic semiconductors due to their compatibility with flexible plastic substrates resulting in lightweight, inexpensive, ergonomic and aesthetic products. While the electrical failure mechanisms in OPVs have been thoroughly investigated, little is known about their mechanical stability, which is as important and critical to ensure long term reliability. The characteristic thin films stresses of each layer present in organic solar cells, in combination with other possible fabrication, handling and operational stresses, provide the mechanical driving force for delamination of weak interfaces or even their decohesion, leading to a loss of device integrity and performance. In this study, we developed a technique to probe weak layers or interfaces in inverted polymer:PC₆₀BM solar cells, establishing a new set-up for the so-called probe tack making it similar to a pull-off test. With this in hand, the adhesion of a variety of active layers towards a standard water-based poly(ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) formulation was compared against that of a new organic-solvent based PEDOT:PSS. The organicsolvent based PEDOT:PSS formulation showed improved adhesion compared to the standard water-based formulation. Chemical characterization of the exposed surfaces indicated a difference in their composition, suggesting a stronger interaction of the organic-solvent based PEDOT:PSS with the active layer and resulting in a different fracture path. Indeed, all OPVs with water-based PEDOT:PSS showed a delamination localized at the active layer/PEDOT:PSS interface, while those with the organic-solvent formulation were not fractured at this interface but rather, in the active layer.

1. Introduction

One of the main factors limiting the commercialization of organic photovoltaic devices (OPVs) is their long-term reliability. So far, most of the research interest has been devoted either to the study of the chemical stability of single materials or blends under different conditions or to the evolution of photovoltaic properties [1]. In particular, the interaction of the different layer in the stack has been studied mostly in terms of diffusion and induced degradation [2]. It has long been known that inorganic PV thin films exposed to tensile and compressive stresses experience increased series resistances, notably due to cracks, as well as to layer delamination [3]. In the case of the organic photovoltaics, the mechanical integrity is determined by several factors [4]. First, the device structure itself (a stack of several thin layers of different types of materials such as metals, oxides and organics) makes the cohesion of the whole device problematic. Second, device

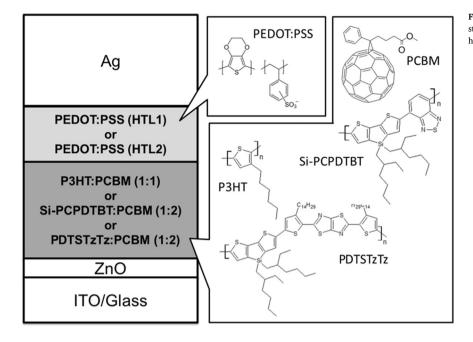
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fabrication induces mechanical stresses such as tensile or compressive strain, which vary with each layer. Third, under operating conditions exposure to the outside atmosphere can induce additional stresses and degradation in different layers which can impact the mechanical integrity of the devices. The synergic effect of these various stresses can lead to a loss of mechanical integrity and eventually to device failure [5].

OPV modules are extremely likely to undergo the same kind of mechanical integrity losses as PV thin film modules. This possibility was foreseen almost 10 years ago [6], based on observations of organic light emitting diode (OLED) degradation [7–13]. Depending on the materials and device architectures used, several degradation mechanisms can participate in the delamination process. When degradation occurs at the active layer (AL)/electrode interface, there is commonly a loss in the interfacial surface area resulting in a reduction of the charge transfer and extraction, and arising from delamination [7], the creation of voids

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Fig. 1. Laver stack of the inverted devices investigated in this study and chemical structure of the different p-type polymers and hole transport layers.

[8], or chemical reactions at the metal electrode surface which form electrically insulating patches [9-11]. More often is the combination of both chemical and physical factors. Diffused water can react at the AL/ electrode weakening the interface, and with long exposure times or when mechanical stresses are present, can lead to delamination [12.13].

Early examples from the literature focused mainly on organic/inorganic interfaces and their property mismatches [8,14]. Na et al. [14] reported on different mechanical stabilities of indium tin oxide (ITO) and ITO-free flexible devices, showing how variations in the nature of the conductive poly(ethylene dioxythiophene):poly(styrene sulfonate) (PEDOT:PSS) and of the ITO (brittle crystalline material) can enhance the mechanical stability of the device. Similarly, Lloyd et al. [8] studied the effect of a PEDOT:PSS hole transporting layer (HTL) in an inverted device architecture under photo-oxidative conditions. Upon aging, the loss of the active area was attributed to delamination. The presence of the HTL strongly reduces this loss and changes its occurrence, which is not evenly distributed as in the case of bare Ag contact, but rather develops from the edges and through pin-holes defects already present in the pristine samples. These few publications only reported the observation of loss of mechanical integrity, without any particular measure of the adhesive properties of the materials or of the layer stack. The area has, however, been explored over the last three years by the Dauskardt group who used two beam-bending techniques, namely, four-point bending and a double-cantilevered beam test. Through these techniques, they investigated the dependence of adhesion/cohesion properties in OPVs on architecture [15], processing [13,16-18] (deposition and post-deposition), materials for the active [19] and hole transporting layer [15], different layer stresses [20] and dump-heat exposure [21] and built specific fracture mechanic models based on the material properties.

This work reports, for the first time, the application of an alternative test, the so-called pull-off test [22], to easily characterize the adhesive strength of an OPV multilayered stack. Its straightforward application and the absence of any sample preparation make it ideal to evaluate the mechanical integrity of such devices, even on-site. The reliability of the technique has been exploited using inverted architecture devices with poly(3-hexylthiophene)/ phenyl-C₆₁-butyric acid methyl ester (P3HT:PC₆₀BM) as the active layer, then extended to two other low band-gap materials, namely poly[2,6-(4,4-bis(2-ethylhexyl)dithieno [3,2-b:2,3-d]silole)-alt-4,7-(2,1,3 benzothiadiazole)] (Si-PCPDTBT) and poly[4,4'-bis(2-ethylhexyl)dithieno[3,2-b:2',3'-d]silole] - 2,6-diyl-alt-[2,5-bis(3-tetradecylthiophen-2-yl)thiazole-[5,4-d]thiazole-1,8-diyl] (PDTSTzTz) in combination with PC₆₀BM. A second study explores the effect of two commercially available PEDOT:PSS formulations, Clevios™ HTL Solar and HTL Solar 2, respectively, with water and organic-nonpolar based solvent. The layer stack of the inverted devices, as well as the chemical structure of the investigated materials, are reported in Fig. 1.

The location of the fracture was determined by analysing the chemical composition by photoelectron spectroscopy, and the topography by atomic force microscopy, of the newly exposed surfaces after the pull-off test. Moreover, the data obtained allowed us to discuss the reasons of this weakest point.

2. Experimental section

2.1. Device fabrication and characterization

The layer stack was based on the model system ITO/ZnO/ Polymer:PC₆₀BM/ PEDOT:PSS/Ag with the following optimized layer thicknesses: ZnO (30 nm), P3HT:PC₆₀BM (1:1) (230 nm), Si-PCPDTBT:PC₆₀BM (1:2) (200 nm) and PDTSTzTz:PC₆₀BM (1:2) (200 nm), PEDOT:PSS with either Clevios[™] HTL Solar (50 nm) or HTL Solar 2 (100 nm), Ag (200 nm). Clevios[™] HTL Solar 2 is a PEDOT:PSS dispersion based on a solvent mixture (mainly 1,2-propanediol, ethanol and diethyleneglycol). The water content is less than 5% [23]. Thermal annealing was performed at different temperatures for different time, both after AL and HTL deposition. The choice of a thick silver electrode (200 nm) was made in order to avoid any interaction between the glue monomers and solvents and the organic layers. The layer thickness was measured with a Stylus Profilometer (Dektac 150, Veeco) on different

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