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Adhesion properties of inverted polymer solarcells: Processing and film structure parameters

Stephanie R. Dupont [a], Eszter Voroshazi [b,c], Paul Heremans [b,c], Reinhold H. Dauskardt [a,*]

[a] Department Materials Science and Engineering, Stanford University, Stanford, CA, USA
[b] IMEC vzw, Leuven, Belgium
[c] ESAT, KULeuven, Leuven, Belgium

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ABSTRACT

We report on the adhesion of weak interfaces in inverted P3HT:PCBM-based polymer solar cells (OPV) with either a conductive polymer, PEDOT:PSS, or a metal oxide, molybdenum trioxide (MoO_3), as the hole transport layer. The PEDOT:PSS OPVs were prepared by spin or spray coating on glass substrates, or slot-die coating on flexible PET substrates. In all cases, we observed adhesive failure at the interface between the P3HT:PCBM with PEDOT:PSS layer. The adhesion energy measured for the solar cells made on glass substrates was about 1.8 J/m^2, but only 0.5 J/m^2 for the roll-to-roll processed flexible solar cells. The adhesion energy was insensitive to the PEDOT:PSS layer thickness in the range of 10–40 nm. A marginal increase in adhesion energy was measured with increased O_2 plasma power. Compared to solution processed PEDOT:PSS, we found that thermally evaporated MoO_3 adheres less to the P3HT:PCBM layer, which we attributed to the reduced mixing at the MoO_3/P3HT:PCBM interface during the thermal evaporation process. Insights into the mechanisms of delamination and the effect of different material properties and processing parameters yield general guidelines for the design of more reliable organic photovoltaic devices.

1. Introduction

Organic photovoltaic (OPV) devices typically involve materials that are compatible with flexible plastic substrates resulting in light-weight and decorative products using inexpensive roll-to-roll (R2R) module fabrication [1–3]. Current state-of-the-art OPVs have reached over 10% power conversion efficiency (PCE), one of the critical milestones for market introduction [4], although they remain limited in size. While the electrical failure mechanisms in OPVs have been extensively investigated [5–7], little is known about their mechanical stability, which is an equally important factor determining their reliability during processing and in service. It is well established that the processing yield and the long-term reliability of multilayer electronic devices are strongly influenced by the adhesive and cohesive properties of internal bi-materials and thin films, respectively [8–10]. Adhesive or cohesive failure may result from thin-film strains and associated elastic stresses present in OPVs [11–13]. Differential strains between the OPV layers may arise from shrinkage during solution processing, thermal expansion mismatch, growth from metallization, mechanical handling including flexing and bending, and other operational and environmental forces, such as wind and hail. The resulting mechanical stresses provide the driving force for delamination of weak interfaces or decohesion of weak layers. This leads to a loss of mechanical integrity and device performance.

In this study, we report on the adhesion of weak interfaces in inverted poly(3-hexylthiophene) and

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* Corresponding author. Address: Department of Materials Science and Engineering, Stanford University, Stanford, CA 94305-2205, USA. Tel.: +1 650 725 0679; fax: +1 650 725 4034.

E-mail address: dauskardt@stanford.edu (R.H. Dauskardt).

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1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C_{61} blend (P3HT:PCBM) based polymer solar cells. The critical adhesion or cohesion energy, G_c (J/m^2), is defined as the macroscopic work of fracture per unit area to separate two layers or break a layer, respectively [8]. This property is typically strongly dependent on the material properties, including chemistry and microstructure, mechanical loading mode, and processing conditions. Within a stack of thin films, the crack will usually propagate along the weakest interface or layer, resulting in adhesive or cohesive failure, respectively. A double cantilever beam (DCB) based adhesion technique (Fig. 1) was used to measure the adhesion energy of the interfaces in the inverted OPV structure. This technique is well established and has the advantage of providing quantitative and reproducible measurements, as demonstrated in previous studies [8,14]. The choice of the inverted device architecture is motivated by its intrinsic air stability and compatibility with R2R processing without compromising device efficiency, proving the industrial relevance of this device architecture.

P3HT:PCBM based OPVs with two types of hole transport layers (HTLs) were selected for study. The first HTL is the well known highly conductive polymer poly(3,4-ethylenedioxythiophene) poly(styrene-sulfonate) (PEDOT:PSS). It is widely used as a HTL and also as a transparent electrode in organic electronic devices because of its high visible light transmission combined with elevated conductivity. Additionally, its formulation can be adapted to a variety of printing techniques without a loss of opto-electronic properties. However, due to its hygroscopic nature, PEDOT:PSS loses its mechanical strength and cohesion in humid environments [15,16] and is also subjected to photo-oxidation [17,18]. From a stability point of view, these are the two main reasons why more stable alternatives like metal oxides are being considered [19–21]. The second HTL selected for study was molybdenum trioxide (MoO_3). This metal oxide HTL was compared with our previously reported study of flexible vanadium oxide (V_2O_5) based solar cells [11].

In order to assess the effect of the processing method, the P3HT:PCBM and PEDOT:PSS layers were prepared by spin or spray coating on glass substrates, or R2R slot-die coating on flexible PET substrates. In all cases, we observed adhesive failure at the interface between the P3HT:PCBM and PEDOT:PSS layers. The adhesion energy of this interface was comparable for all the solar cells made on glass substrates, but was lower for the R2R processed flexible solar cells. The adhesion energy was independent of the PEDOT:PSS thickness in the range of 10–40 nm. A marginal increase in adhesion energy was measured with increased O_2 plasma power. Compared to solution processed PEDOT:PSS, we found that thermally evaporated MoO_3 adheres less to the P3HT:PCBM layer. This was most probably due to the reduced mixing with P3HT:PCBM during the thermal evaporation process. Finally, a number of guidelines to improve the fracture properties of P3HT:PCBM-based OPVs will be suggested.

2. Experimental

2.1. Solar cell preparation

The structure of the inverted P3HT:PCBM-based polymer solar cells is shown in Fig. 1. A zinc oxide (ZnO) precursor was deposited from solution onto cleaned indium tin oxide (ITO) coated glass substrates (Kintec). A 250 nm active layer composed of 1:1 wt.% mixture of P3HT:PCBM was deposited from solution. This solution contained as purchased P3HT and PCBM dissolved in o-dichloro-benzene (oDCB) to spin and spray coat the P3HT:PCBM layer, as summarized in Table 1. 1,3,5-trimethylbenzene (mesitylene) was added to the solution for spray coating to favor substrate coverage and reduce surface roughness. Note that in the case of the R2R processed flexible OPVs, P3HT and PCBM were dissolved in chloro-benzene (CB) to slot-die coat the P3HT:PCBM layer. Prior to thermal evaporation of the silver electrode (Ag, 100 nm), a thin HTL was deposited. The HTL was either a solution processed PEDOT:PSS layer or a thermally evaporated 30 nm MoO_3 layer. The PEDOT:PSS layer was deposited from a commercially available water-based dispersion (Baytron PVP AI 4083 or EL-P 5010) selectively diluted by isopropanol (IPA) and deionized water (DIW) as summarized in Table 1. Spin coated PEDOT:PSS layers with different thicknesses were deposited by systematically varying the spin coating velocity. A 60 s oxygen (O_2) plasma treatment on the spin and spray coated P3HT:PCBM layers prior to PEDOT:PSS deposition was used to enhance the wettability of PEDOT:PSS solution with the plasma power (kW), the spin coating velocity (rpm) and corresponding layer thickness (nm) reported in Table 2. Further details on the processing on glass substrates [18,19] and the R2R processing on flexible polyethylene terephthalate substrates [20] have been previously reported.

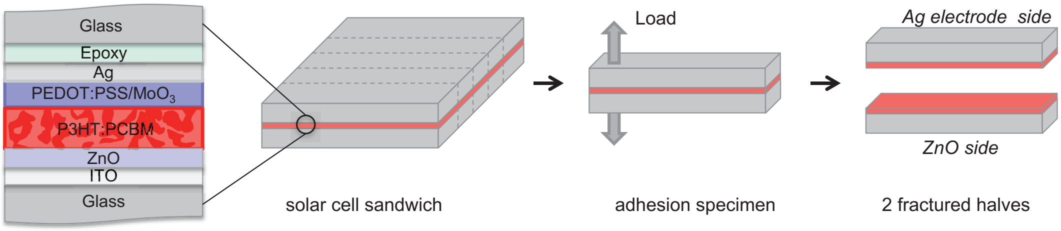

Fig. 1. Illustration of the solar cell structure and the square glass sandwich. With a wafer saw, 2 identical DCB adhesion specimens are cut out. After adhesion testing, 2 fractured halves are obtained and referred to as the Ag electrode side and the ZnO side.