



Electrode de-wetting as a failure mechanism in thermally-aged OPV devices

William Greenbank, Lionel Hirsch, Sylvain Chambon*

Univ. Bordeaux, IMS, CNRS, UMR 5218, Bordeaux INP, ENSCBP, F-33405 Talence, France



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ABSTRACT

Organic photovoltaic (OPV) solar cells are a promising solar energy technology that could greatly increase the number of ways that solar cells can be utilised. Many of these applications involve the use of thin silver electrodes, however little attention has been paid to the role that electrode thickness plays in the operational stability of the device. This study found that inverted OPV devices with thin silver electrodes exhibited a catastrophic failure with only modest amounts of thermal ageing – primarily driven by substantial increases in the series resistance and decreases in the fill factor of the devices. Subsequent analysis by atomic force microscopy and Rutherford backscattering spectrometry showed that as the devices aged, gaps in thin silver electrodes appeared, showing that the electrode was de-wetting from the device, which would explain the rise in series resistance. This represents an intrinsic vulnerability in inverted OPV devices that can be avoided if the silver electrode is sufficiently thick (> 60 nm). This problem can be overcome with the addition of a thin metal-oxide layer on top of the electrode which greatly slowed the electrode de-wetting with little variation in device performance – representing a potential solution for applications where a thin silver electrode is required.

1. Introduction

Organic photovoltaic (OPV) solar cells are a third-generation photovoltaic technology that has the potential to help meet the world's rising demands for energy without adding to greenhouse emissions [1,2], as well as open up new applications for photovoltaic solar energy [3]. Their key benefit is that they can be printed onto flexible substrates at room temperature – a process that is both low cost and highly scalable [4,5]. The principal barriers to their commercialisation have, thus far, been their low power conversion efficiencies (PCE) as compared to their inorganic counterparts, and their short lifetimes [6,7]. With respect to PCE, significant progress has been made, and it is now estimated that OPV modules would be commercially competitive with silicon modules once production is scaled up [8]. However, for this to be true, significant improvements in device lifetime are needed.

OPV device degradation is typically driven by four principal stimuli: oxygen, water, heat and light (mechanical stress can also play a role in certain applications) [9]. These stimuli can be roughly split into intrinsic stimuli and extrinsic stimuli. An extrinsic stimulus is one that can be removed without impeding the functioning of the OPV device, such as oxygen and water, which can be removed through the use of encapsulation. On the other hand, intrinsic stimuli are always present during the normal operation of a device and, as such, degradation pathways involving intrinsic stimuli can only be avoided or minimised through device design and the choice of materials.

An example of an intrinsic degradation stimulus is heat. Thermal degradation has long been known to play a significant role in the loss of OPV device performance, particularly in devices containing bulk heterojunction (BHJ) active layers [10]. In such devices, the morphology of the BHJ can be altered when the device is held at high temperatures, resulting in the growth of donor and acceptor domains beyond the exciton diffusion length, increasing the geminate recombination rate, and thereby resulting in a loss of short-circuit current density (J_{SC}) [11,12]. This form of thermal degradation can be largely avoided through the use of a high-glass transition temperature (T_g) polymer donors [13], or through post-deposition crosslinking [11]. In a previous study, we demonstrated that thermal degradation can also give rise to a loss of open-circuit voltage (V_{OC}) when MoO_3 is used for the hole transport layer (HTL) and silver is used for the top electrode in inverted OPV devices [14]. It was proposed that this behaviour was due to the diffusion of species from the top layers of the device into the active layer, resulting in the formation of charge traps and an increase in the monomolecular recombination rate. A subsequent study demonstrated that interdiffusion does occur when a silver electrode is used, and that this also has consequences for the mechanical stability of the device [15].

We also reported in this previous study that thermal ageing of OPV devices containing silver electrodes gives rise to significant changes in the surface morphology of the top electrode, specifically a significant increase in the surface roughness [14]. This roughness increase was

* Corresponding author.

E-mail address: sylvain.chambon@ims-bordeaux.fr (S. Chambon).

significant, but not comparable to the thickness of the electrodes used in the devices, and therefore did not appear to have any effect on the performance of the devices. It was not clear, however, what the effects would be on a device with a thinner silver electrode. In the pursuit of replacements for indium tin oxide (ITO) as a transparent electrode, many studies report the use of thin layers of silver or silver nanowires [16–20], however little is known about their resilience to degradation stimuli when incorporated into OPV devices. Therefore, this study explores the effects that thermal degradation can have on thin silver electrodes, both with respect to physical alterations to the electrode and to changes in the performance of OPV devices with these electrodes. OPV devices with electrodes of different thicknesses were thermally aged and the changes in their key photovoltaic parameters were examined. Rutherford backscattering spectrometry (RBS) and atomic force microscopy (AFM) experiments were employed to observe physical changes in the electrode with thermal ageing. All ageing was carried out under ISOS-D-2 conditions [21]. By comparing the results of the two parts of the study, silver electrode thickness-dependent sensitivity to thermal ageing was able to be correlated to physical changes in the electrode – a result which can inform the future design of silver-containing OPV devices.

2. Methods

The RBS samples were measured on a beamline of a 2 MeV He^+ particle accelerator at the Bordeaux-Gradignan Centre for Nuclear Research (*Centre Etudes Nucléaires de Bordeaux Gradignan*) with an ion current of approximately 1 nA, beam diameter of 3 mm, and a scattering angle of 160° . The samples were layers of HTL oxide (WO_3) and electrode metal (aluminium or silver) deposited on glassy carbon substrates (Fig. 1(a)), using e-beam and thermal evaporation respectively [14]. The layer thicknesses used are given in Fig. 1(a). Each material combination had a control sample that was stored in a glovebox under an inert atmosphere (N_2 atmosphere: < 0.1 ppm O_2 , H_2O), and a sample that was thermally aged at 85°C in the dark, in a glovebox under an inert atmosphere.

The AFM measurements were carried out with a Bruker Dimension FastScan atomic force microscope in tapping mode using silicon cantilevers (Asylum Research AC-160-TS) with a typical tip radius of ≈ 7 nm, a spring constant of 26 N m^{-1} and a cantilever resonance frequency of approximately 300 kHz.

The surface energies of the different surfaces were determined using a Krüss DSA 100 goniometer by the method proposed by Owens and Wendt [22] based on the contact angle measurements of different liquids on the substrate surface at 20°C in static mode. The results correspond to the mean of at least 3 measurements.

The OPV devices (Fig. 1(b)) were fabricated in the inverted architecture with poly-(3-hexylthiophene):[6,6]-phenyl- C_{61} -butyric acid methyl ester (P3HT:PCBM) BHJ active layers. The ZnO layer was deposited on ITO/glass substrates by spin coating with a zinc(II) acetate sol gel [23], and subsequently annealing for 30 min at 180°C in air. The samples were then transferred to a glovebox (N_2 atmosphere: < 0.1 ppm H_2O , O_2), in which all subsequent fabrication, characterisation, and ageing steps were carried out. The active layer (1:1 w/w P3HT:PCBM BHJ blend) was then deposited by spin coating the samples with a filtered solution containing 20 mg mL^{-1} P3HT (Plextronics Plexcore OS2100, 53 kDa) and 20 mg mL^{-1} PCBM (Solaris Chem.) in 1,2-dichlorobenzene. The active layers were annealed in saturated 1,2-dichlorobenzene vapour to attain optimal BHJ morphology. The WO_3 and silver layers were subsequently deposited by e-beam and thermal evaporation respectively. The thicknesses of the layers were measured by either mechanical profilometry (KLA-Tencor, Alpha-Step IQ) for the solution-processed layers (ZnO and BHJ), or by resonating quartz balance during deposition for the layers deposited by evaporation. All device ageing was carried out at 85°C on a covered hot plate in a glovebox for up to 1100 h while periodically characterised by IV analysis (AM1.5 HMI solar simulator set at 1000 W m^{-2} with a calibrated radiometer (IL 1400 BL) – the samples remained in a glovebox for measurement).

3. Results and discussion

3.1. Thermal ageing of thin electrode OPV devices

Inverted OPV devices with varying-thickness silver electrodes were prepared with the structure shown in Fig. 1(b), and thermally aged for up to 1100 h at 85°C while being regularly characterised by IV measurement. The results from these experiments are presented in Fig. 2, and the initial device PV statistics in Table 1.

As shown in Fig. 2(a), the sample group with 20 nm thick electrodes exhibits a catastrophic failure in which most of the devices have failed by 50 h of thermal ageing, whereas the other sample groups do not. The device failure is driven initially by a decline in FF which is followed by a decline in J_{SC} (Fig. 2(c)).

Focusing on the devices with 20 nm thick electrodes, the change of the IV curve during the device failure is shown in Fig. 2(d). It is evident from these results that there is a significant increase in the series resistance in this device, decreasing the fill factor and, as it becomes more severe, eventually suppressing all photocurrent. As shown in Fig. 2(b), across all the 20 nm devices there is a very sharp rise in the series resistance that begins around 20 h of thermal ageing. Increasing the electrode thickness leads to an increase of the devices stability. With a

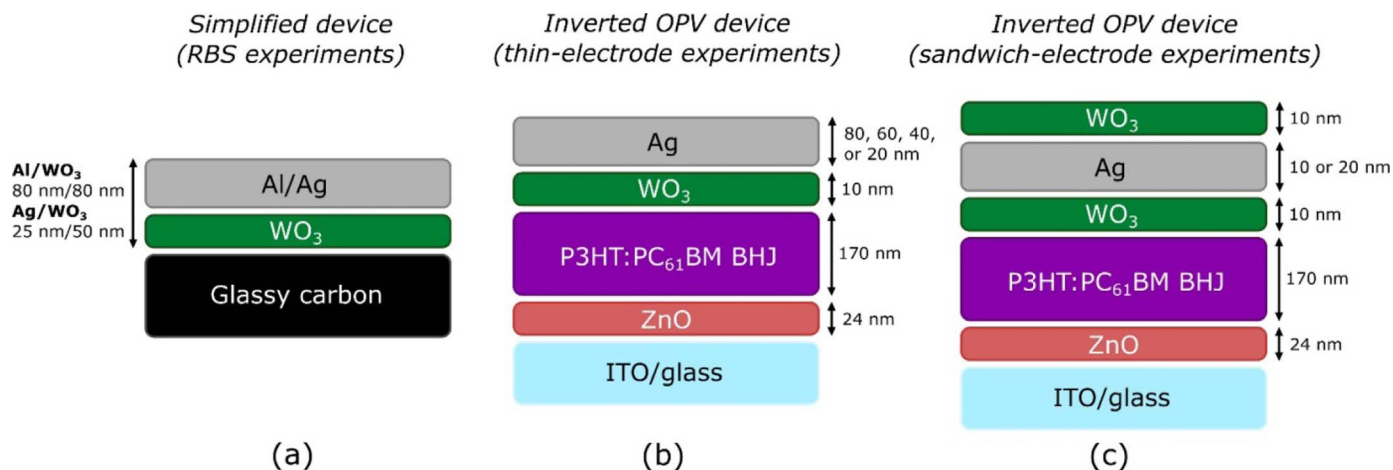


Fig. 1. The structures and layer thicknesses of the samples used in this study for: (a) the RBS experiments (Section 3.2), (b) the IV studies of thin silver electrode OPV devices (Section 3.1), and (c) the IV studies of OPV devices with the higher stability sandwich electrode (Section 3.3).

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