



Blue photon management by inhouse grown ZnO:Al cathode for enhanced photostability in polymer solar cells



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ABSTRACT

We report the improvement in photostability of P3HT:PC₆₀BM based bulk heterojunction solar cells deposited on Al-doped ZnO as a cathode layer replacing ITO as regularly used TCO in cells with N-I-P configuration. We experimentally and theoretically demonstrate that use of thicker ZnO:Al as cathode can successfully cut down the rate of photodegradation in short circuit current by ~40% and open circuit voltage by ~30% compared to the control device made on ITO based cathode. This effective reduction in photodegradation is understood to be coming from the absorption of ultraviolet and blue photon in the cathode layer itself. The loss in short circuit current due to the loss of blue photon in EQE is compensated by higher FF (lower series resistance) due to thicker ZnO:Al layer resulting in final device efficiency almost uncompromised with added benefit of reduced photo degradation. The experimental results are supported with optical simulations which show more absorption in the short wavelength region for the thicker ZnO films, compared to ITO films, deposited on glass substrates. This work also proposes using ZnO:Al cathode as a template for random textured front surface to potentially increase short circuit current by increase in photon absorption in active layer matrix by light scattering techniques. Our results provide an inexpensive pathway for improving the stability of organic photovoltaics without compromising the device performance.

1. Introduction

Recently there has been a considerable interest in organic material based solar cells due to their flexibility, convenience in tunability of their opto-electronic properties etc. However various types of degradation both intrinsic [1–11] and extrinsic [12–15], present considerable amount of challenge to the complete realization in harnessing the full potential of such materials. There was considerable effort invested already to study, understand and engineer mechanisms by which such degradation effects can be eliminated or to the best can be reduced considerably. It was proposed that organic material based solar cells are prone to light induced degradation even under inert atmosphere mainly due to the atomistic migration of either H or O within the polymer cluster along the backbone [9]. Such degradation was found to create electrically active mid-gap defect states shown in Fig. 1A, which acts as a center of charge carrier recombination and generation which happen to be located at the interface of donor-acceptor and they are shown to be energetically located around the mid-gap (0.55–0.6 eV) of the

interface band gap. It is also noted that such defect states are metastable in nature and can be recovered with conjunction of appropriate device architecture and post degradation thermal annealing. [9,10,16]

There are numerous attempts to eliminate or reduce the amount of photo-degradation which includes the use of inorganic amorphous silicon as electron transport layer (ETL) [17], use of amorphous silicon filter, amorphous silicon-organic (inorganic/organic) tandem solar cells etc. [18–20]. The motivation behind all such work is to reduce the amount of ultraviolet and blue photon in the range of 350–500 nm which has sufficient energy to initiate the atomic migration of H from C–H bond [9,16,21] in the aromatic chain resulting in the creation of interface defect states. Amorphous silicon, being a wide band gap material has capacity to absorb ultraviolet and blue photon while leaving the lower energy photon for active layer absorption and consequently photo-carrier generation. However, such efforts inherently involve complex process of PECVD high vacuum deposition of amorphous film. In addition, often the idea of using only a filter of a:Si-H reduces blue photon absorption and thus reduces short circuit current unless it can

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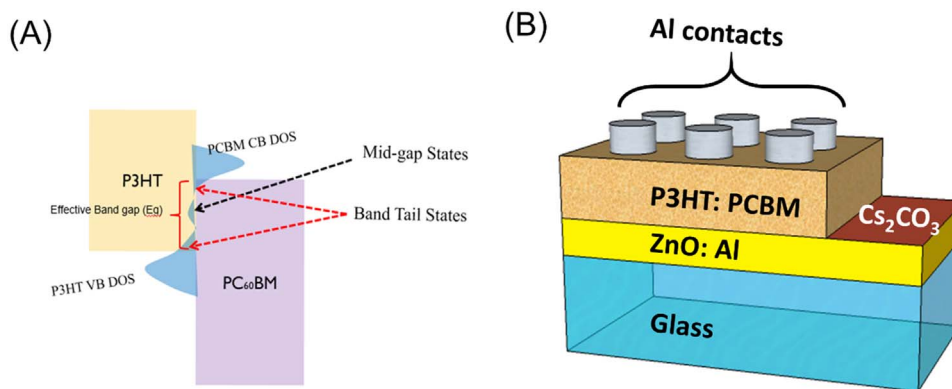


Fig. 1. (A) Schematic showing energy location of mid-gap defect states. (B) Layer wise schematic of an (N-I-P) solar cell on ZnO:Al substrate.

be used as a top cell for inorganic/ organic tandem solar cells.

Here, we grew customized ZnO:Al by RF magnetron sputtering on regular glass substrates without any commercial ITO on them for fabricating organic solar cell as shown in Fig. 1B. There are several motivations for using ZnO:Al as a transparent conductive cathode for organic photovoltaic application. Indium based TCOs are widely used as transparent conductive electrodes for solar cell as well as LED applications since beginning of the research in this field. However, Indium is a rare earth material and not available in abundance in the earth crust [22–24]. In addition, ITO has an environment concern as mentioned in some literature [25]. ZnO:Al, on the contrary is widely available material and very much cost effective in comparison to conventional ITO based TCO [26]. Next ZnO:Al can be easily deposited by RF magnetron sputtering with optimized electrical and optical property [27–30]. Finally thicker ZnO:Al TCO can also be etched easily by dipping the film coated on glass in diluted HCl as per etching recipe reported elsewhere [31], to obtain random roughened structure which can be used as both front and back end reflector that can scatter and rebound the incoming light resulting in increasing the light path inside the active medium of solar cell [32,33].

ZnO:Al has previously been used as a cathode for ITO-free inverted polymer solar cells. However, the focus was to demonstrate improvement in efficiency from better transmittance of ZnO:Al cathode in the absorption band ($\lambda \sim 500\text{--}600\text{ nm}$) of organic solar cells [34]. In the present work, we experimentally and theoretically study the effective optimization of ZnO:Al cathode thickness to improve the photostability of organic solar cells without compromising the efficiency. We show the results of P3HT:PC₆₀BM bulk heterojunction devices that were fabricated on optimized substrate coated with ZnO:Al cathode. We report the device results of four different N-I-P (*inverted*) bulk heterojunction devices fabricated on three different ZnO:Al film of different thickness and one control device prepared on regular ITO coated glass substrate. We selected the best performing device out of the batch and performed the photodegradation testing. The device with optimized (thickness = 1.25 μm) cathode film shows almost 40% reduction in the rate of degradation of in-situ measured photo current.

2. Results and discussion

2.1. ZnO:Al film fabrication and characterization

The electrical and optical properties of ZnO:Al have been widely studied by various research groups in the past. It is well-known that the increase in substrate temperature would result in increase of ZnO crystal size, resulting in enhanced charge carrier mobility. There are other ways to improve the crystal size of ZnO:Al thin films such as increasing RF power or sputter chamber pressure with minimum drop in transmission in visible spectrum [35,36]. Higher RF power results in high deposition rate for a given temperature and sputter pressure,

leading to the thicker ZnO:Al film. It has been shown that the intensity of the key XRD peak of ZnO:Al at $2\theta = 34.3^\circ$ ((0 0 2) crystal plane) increases and becomes sharper as the film thickness increases, suggesting higher crystallinity. Similarly, for a given RF power and substrate temperature, higher sputter pressure increases films conductivity due to higher carrier mobility and carrier density with thicker film for a given time of deposition [36]. Finally, higher RF power was reported to yield lower sheet resistance for ZnO film without much degradation in visible spectrum transmittance [37]. Our initial objective was to obtain ZnO:Al film that has very high conductivity, transmission at visible spectrum comparable to regular ITO film and lower transmission at the wavelength between 400 nm and 500 nm which are typically the major source of photo-induced degradation in organic solar cells. Such film characteristics can only be obtained with a thicker ZnO:Al film with higher grain size and better crystallinity resulting in lower sheet resistance which is a prerequisite for photovoltaic application.

In our first step of optimization for deposition of ZnO:Al film, we used 2.5mTorr Argon pressure, substrate temperature of 150 C and varied RF power from 80 to 120 W. The deposition was done for a constant time of 10.5 min. The transmission of all the ZnO:Al films were measured by Cary Spectrometer from 400 nm to 800 nm as shown in Fig. 2. The sheet resistances were measured using four-point probe technique and are reported in Table 1. As we can see, increasing RF power actually increases the deposition rate and thus we were able to obtain physically thicker film for a constant deposition time. With an obvious benefit of achieving lower sheet resistance at higher RF power, optical transmission was reasonably comparable to the ITO coated substrate which has a sheet resistance of roughly 16 Ω/square . On an

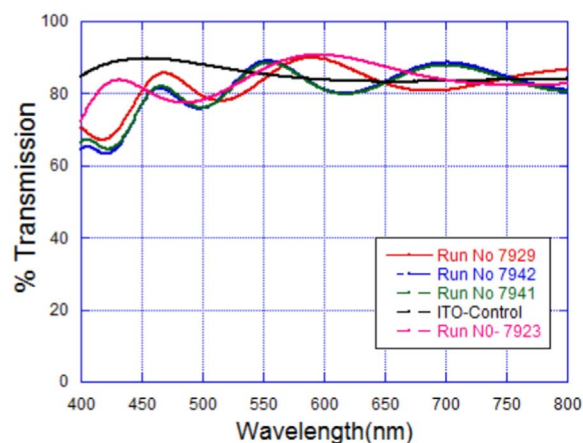


Fig. 2. Transmission characteristics of respective ZnO:Al films deposited and reported in Table 1. The deposition was done for varying RF power and constant time of 10.5 min. The other deposition conditions were not changed (2.5mTorr Argon pressure and 150 C substrate temperature).

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