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Highly transparent micro-patterned protective coatings on polyethylene terephthalate for flexible solar cell applications

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ABSTRACT

Degradation of the performance and efficiency of organic solar cells (OSC) due to surface damage, caused by exposure to harsh environments, continues to be a major problem. Therefore, protective coatings must be developed that minimize damage to the outer surface of OSCs, without compromising their transparency and flexibility. For this purpose, ultra-thin, micro-patterned, single/bi-layer coatings comprised of H-free diamond-like-carbon (DLC) and indium tin oxide (ITO) were fabricated. The coatings were deposited onto polyethylene terephthalate (PET) substrates, using magnetron sputtering. The goal of the micro-patterning was to improve the wear resistance of the coatings, while maintaining the high optical transparency of PET. Optical properties, wear resistance, erosion resistance, and flexibility of the coatings were investigated. Furthermore, the performance of a pre-fabricated OSC with and without the proposed protective coating before and after the wear and erosion tests was investigated. The experimental results demonstrated that the wear resistance and transparency (of the single layer micro-patterned DLC coating), was superior to both bi-layer and continuous DLC coatings. The micro-patterned DLC coating caused very little change in transparency (less than 1.7% reduction in the visible spectrum), and reduced the wear rate (mm^3/N·mm) of PET by ~75%. The erosion resistance of the PET was improved, and its degree of flexibility was unaffected. Overall, the results of OSC performance analysis demonstrated the functionality and potential application of the proposed micro-patterned protective coating for flexible solar cells.

1. Introduction

Global attempts toward replacement of conventional fossil energy sources with green and eco-friendly alternatives have been increasing steadily over the past decade. Solar energy has shown great potential as a viable alternative form of energy, as it is: inexhaustible, sustainable, free, and renewable. Therefore, efforts have been made to develop various types of solar cells (organic and inorganic) with high efficiency and low manufacturing costs. Despite having a lower efficiency than inorganic (silicon-based) solar cells, organic solar cells (OSC) are widely used due to their unique advantages such as cost effectiveness, flexibility, and wearability (Timothy et al., 2016; Park, 2016).

One of the challenges that is as equally important as the inherent efficiency of solar cells, is maintaining their efficiency for prolonged operation in maintenance-free environments (Al Shehri et al., 2016; Humoond et al., 2017; Corazza et al., 2015). The degradation of stability and efficiency in solar cells due to external factors (e.g. water penetration, dust accumulation, wear, and sand erosion), was reported in multiple studies (Jorgensen et al., 2008; Landis, 1996; Bouaouadja et al., 2000). Of these external factors, wear and erosion can physically damage the photovoltaic cells. The physical damage can either entirely compromise the cells inner structure, disrupting the photovoltaic effect, or occur only on the top cover in which case the surface roughness can be altered drastically. Experimental analysis has demonstrated that transparency, sunlight transmission, and overall efficiency decreases exponentially as the surface roughness increases (Kim and Kim, 2012). Therefore, high durability and reliability of the top cover is an important factor in determining the stability of solar cells, when subjected to long periods of exposure in severe environmental conditions. In particular, this issue becomes more serious in the case of OSCs, where only a soft polymer top cover (e.g. polyethylene terephthalate (PET)) is mainly used to protect the inner structure of the cell.

Surface engineering methods that involve manipulation of the micro/nano structure and properties of surfaces, have been widely

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utilized to minimize degradation of system efficiency due to physical damage such as wear and erosion. A widely used method is the deposition of protective coatings. In general, an ideal protective coating must simultaneously fulfill a range of properties including good adhesion to the substrate, high level of hardness, elasticity, low internal stress during deposition, chemical inertness and low coefficient of friction (Penkov et al., 2015; Khadem et al., 2017). A unique material such as Diamond-like-carbon (DLC) not only possesses most of these properties, but also has high transparency in both the visible and infrared wavelength range (Khadem et al., 2014; Umeno and Adhikary, 2005). These properties make DLC a potential candidate for optoelectronic applications. DLC consists of a network of sp^2 and sp^3 hybrids, which generally fall into two categories (hydrogenated and hydrogen-free). In the case of hydrogenated DLC, the optical transparency, and band gap, can be tuned by controlling the hydrogen content (Choi et al., 2008). Hydrogenated DLC has been widely used as an anti-reflective coating on Si-based photovoltaic cells (Kumar Raut et al., 2011; Zhu et al., 2009). Although the optical properties of hydrogenated DLC coatings are tunable, their mechanical properties such as hardness are lower than hydrogen-free DLC coatings, which lowers their wear and erosion resistance. Moreover, the wear and erosion resistance of hydrogenated DLC coatings is sensitive to environmental changes (e.g. humidity and temperature), which limits their practical application as a protective coating in certain applications (Khadem et al., 2014; Sedlacek et al., 2008). Therefore, hydrogen-free DLC coatings are preferable in situations where protection against wear and erosion is desired.

Another well-known method for reducing wear is surface micro/nano-patterning which works mainly due to two mechanisms: reducing the contact area and trapping the wear debris (Tang et al., 2013). When accumulated, the sharp morphology of wear debris can accelerate the wear process, by inducing a high stress concentration at the contact interface (Yue and Wahab, 2016). Surface patterning has also been widely employed in optoelectronic devices to trap light wave and to minimize reflections (Schuster, 2017; Tang et al., 2014). In the case of PET-based OSCs, a variety of methods such as interlayer lithography and direct laser interference patterning (DLIP) have been used for the fabrication of micro/nano patterns (Huang et al., 2017; Meskamp et al., 2012). In a particular study (Meskamp et al., 2012), DLIP was used to fabricate hexagonal periodic patterns on PET to enhance the performance of ZnPc:C_{60} solar cells, by trapping and concentrating the light in the absorber layer. Using this method, it was found that the power conversion efficiency increased by 21%, compared to a flat PET without a surface pattern.

When protecting PET-based OSCs, by applying the described surface engineering methods (coating and patterning) is concerned, maintaining the high transparency of PET is an important requirement. A patterned coating not only maintains the high transparency of the PET (due to coating discontinuity, which results in less surface coverage area), but also reduces wear and surface damage, through the previously discussed mechanisms.

In this study, we report on the development of highly transparent, micro-patterned, wear/erosion protective coatings. The coatings consisted of DLC and indium tin oxide (ITO), which were deposited in single and bi-layer forms using magnetron sputtering. DLC and ITO were selected because of their high optical transparency and good adhesion to the PET substrate. The deposition parameters of DLC were optimized to achieve the lowest amount of wear. The micro-pattern on the coating not only increased the wear resistance but also aided in maintaining the inherent transparency of PET. According to the experimental results, the level of protection of the PET substrate was significantly improved, without any major effect on the light transmission properties and performance of the OSC. Furthermore, the inherent flexibility of the PET substrates remained unchanged.

2. Experimental details

2.1. Coating deposition

Initially the PET substrates were cleaned using ultra-sonication in acetone, then ethanol, and finally di-ionized water, for 5 min each. To improve the adhesion of the coatings, and to enhance the surface energy, the substrates were treated using argon plasma (5 min – 10 W) (France and Short, 1997). The coatings were deposited using magnetron sputtering. Graphite and ITO targets, of 99.99% purity, were used. The initial pressure, prior to deposition, was controlled at 5×10^{-6} Torr. The working pressure was controlled at 2×10^{-3} Torr. DLC and ITO coatings were deposited using a DC power source. Current and frequency of the DC power were set to 250 mA and 40 KHz, respectively. The effect of applied negative bias, as a deposition parameter that significantly affects the mechanical properties of the DLC, was also investigated. For this purpose, three values of negative biases (0, 40 and 70 V) were applied during the DLC deposition.

The micro-patterns were fabricated through masking, using a commercial metal mesh with an opening of ~20 µm. A metal mesh with a small opening size was chosen to minimize the surface coverage area, and therefore, maximize the transparency of the coating. To minimize the gap between the mask and substrate, the metal mesh was fastened tightly to the PET substrate using a custom-made mask holder. Two sets of micro-patterned coatings were prepared. One was a single layer of DLC (5 nm), and the other was a bi-layer composed of ITO (2 nm) and DLC (5 nm). A 5 nm continuous DLC coating was also prepared as a reference, for comparison purposes.

Fig. 1 shows a 3D schematic view of the fabrication process, together with the laser-enhanced optical view of the metal mesh and the fabricated micro-patterns on the PET. Distinct square-shaped micro-patterns, with a dimension of ~20×20 µm, are clearly visible. It was decided that the overall thickness of all the coatings should be less than 10 nm, to maintain the high transparency and flexibility of the PET. The detailed experimental procedure for transparency, wear, erosion and bending tests are described below. All the experiments were performed in a class 100 clean room, and were repeated at least 3 times to ensure the reliability and repeatability of the data.

2.2. Structure and property evaluation

The nano-structure and chemical state of the deposited DLC coating were evaluated, using both X-ray photoelectron spectroscopy (XPS; Thermo UK K-alpha) and Raman spectroscopy (Raman; Thermo Scientific) equipped with a Nd:YAG laser of wavelength 532 nm. These analyses are particularly important for investigating the sp^2 and sp^3 hybrids, because the properties of DLC coatings are strongly dependent on the ratio of sp^3 to sp^2.

The hardness and internal stress of DLC coatings, deposited at different negative biases, were measured using a high precision ultra nano-indenter (CSM; UNHT), and a surface profiling system (Bruker; DekTek), respectively. For this purpose, different sets of DLC coatings, with sufficient thicknesses, were prepared on Si substrates. The indentations were performed using a Berkovich tip with a diameter of 100 nm. The maximum indentation depth was less than 10% of the DLC coating thickness, to avoid the substrate effect.

2.3. Light transmission analysis

The transparency of the coatings was evaluated using a UV/VIS spectrophotometer (V-650; JASCO), over the visible light wavelength range (400–800 nm), and compared with the transparency of bare PET substrate. The laser spot size was 100 µm, and the scan speed was 1000 nm/min.

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