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## Photodegradation of small-molecule organic photovoltaics

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## ABSTRACT

We investigate the photostability of organic photovoltaic (OPV) cell active layers comprised of the archetype donor, boron-subphthalocyanine chloride (SubPc), and fullerene acceptors, aged under either AM1.5G illumination or in the dark, and in either air or inert atmosphere. Under long-term exposure to light, we observe significant photobleaching and crystallization of SubPc. Mixing SubPc with C<sub>60</sub> as is commonly done in high efficiency small molecule OPVs, the crystallite formation is inhibited and the bleaching is suppressed due to a significantly reduced exciton lifetime in the blends. Furthermore, the spectral dependence of the degradation suggests that photo-dimerization of C<sub>60</sub> is an important factor leading to burn-in loss in efficiency previously reported in SubPc/C<sub>60</sub> OPVs. The existence of dimerization is supported by Fourier transform infrared spectroscopy data taken both before and after exposure to light. Increasing the fraction of SubPc in a SubPc:C<sub>60</sub> blend leads to a decrease in the rate of film degradation, providing further evidence for C<sub>60</sub> dimerization. Due to its reduced tendency for photo-dimerization, C<sub>70</sub> is more stable than C<sub>60</sub> when used in small molecule OPVs.

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## 1. Introduction

High efficiency, long lifetime, and low cost manufacturing are the three pillars of a successful solar cell technology. Among the various emerging technologies, organic photovoltaic cells (OPVs) are a promising option for renewable energy due to their potential for flexible, low-cost, and large-scale production. While the efficiency of organic photovoltaics is rapidly approaching that required for commercialization [1], their operational lifetimes are below approximately 25 years that is deemed acceptable for practical deployment [2].

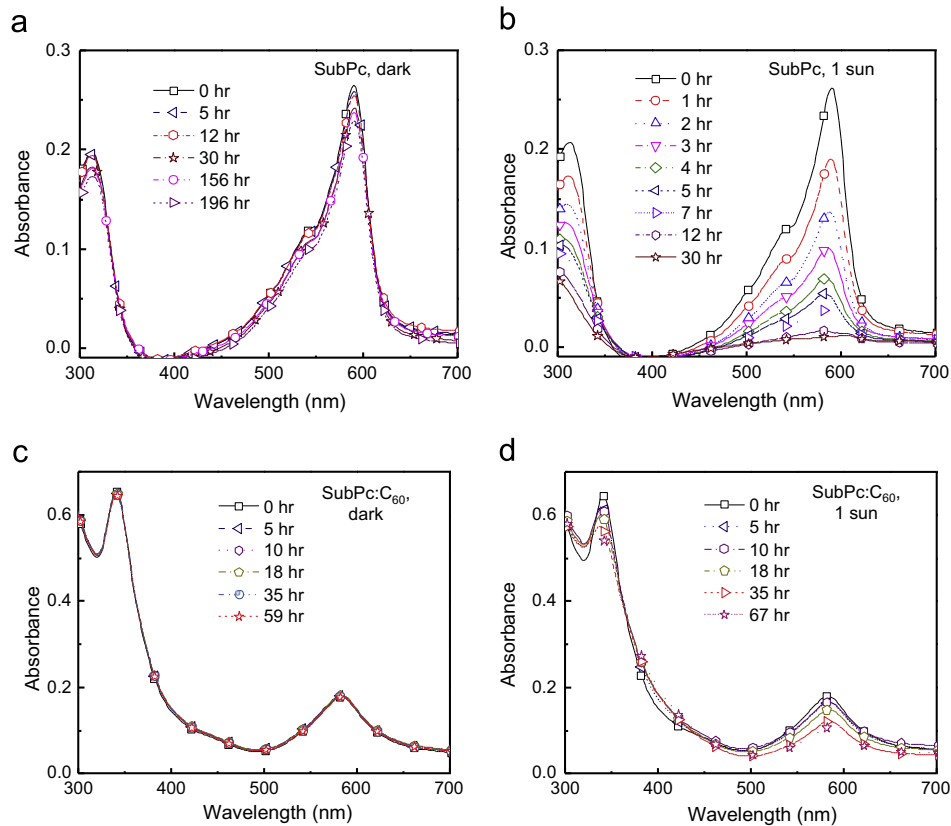
Degradation in OPVs can be caused by deterioration of organic molecules [3–9], oxidation of electrodes [10], delamination and reaction of the organic layer with the electrode [11–13], etc. The intrinsic stability of the organic active layer is dependent on both the materials and device structures used. Organic thin film degradation may occur via exciton-induced defects [14–16], morphological changes, photo-oxidation, and other photochemical and thermochemical reactions [4,6–8,17,18]. In polymer solar cells, photo-oxidation in the donors, poly[2-methoxy-5-(2'-ethylhexyloxy)-p-phenylenevinylene] (MEH-PPV), poly[2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylene vinylene] (MDMO-PPV) and poly(3-hexylthiophene)

(P3HT), has been observed to lead to film bleaching [5,6,19,17]. In addition, the fullerenes are the most commonly used acceptor materials in high efficiency OPVs due to their large absorption coefficients, long diffusion lengths, and high electron mobilities. However, C<sub>60</sub> and the substituted fullerene ([6,6]-phenyl-C<sub>61</sub>-butyric acid methyl ester, PC<sub>60</sub>BM) are known to polymerize upon irradiation [20,21]. In addition, Distler et al. showed that dimerization of PC<sub>60</sub>BM leads to the reduction of fill factor and short circuit current in an OPV cell [21]. However, to the best of our knowledge, there have been no reports of the effects of C<sub>60</sub> dimerization on device performance.

Previously, we found that exciton-induced traps in C<sub>60</sub> are responsible for the deterioration in power conversion efficiency in fullerene-based small molecule OPVs during early stage burn-in [16]. In this work, we examine the photostability of boron subphthalocyanine chloride (SubPc) and C<sub>60</sub> thin films employed in that initial study, and find that the burn-in degradation is due to photoinduced dimerization along with photobleaching of the SubPc. Adding either C<sub>60</sub> or C<sub>70</sub> to the SubPc to form a mixed donor/acceptor active region greatly reduces photo-degradation by facilitating rapid electron transfer to the acceptor before the high energy excitons can cause chemical reactions in the photo-active molecules that result in trap formation. Furthermore, the use of a C<sub>70</sub> acceptor results in more stable OPVs due to its reduced tendency to photo-polymerize.

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**Fig. 1.** Time evolution of absorbance spectra in air for (a) boron-subphthalocyanine chloride (SubPc), kept in the dark, (b) SubPc, under 1 sun illumination, (c) SubPc:C<sub>60</sub> (1:4), kept in the dark, (d) SubPc:C<sub>60</sub> (1:4) under 1 sun illumination.

This paper is organized as follows: in [Section 2](#), we present experimental methods, in [Section 3](#) we provide results, and in [Section 4](#) we discuss the physical origins of photobleaching and OPV degradation. In [Section 5](#) we present conclusions.

## 2. Experimental

The SubPc and fullerene films were deposited on Si or ITO-coated glass substrates that were previously solvent cleaned followed by CO<sub>2</sub> snow cleaning using procedures described previously [22]. Following ultraviolet (UV)-ozone treatment, substrates were loaded into a high-vacuum (base pressure  $< 4 \times 10^{-7}$  Torr) thermal evaporation chamber where organic materials were deposited at a rate of 1 Å/s. All deposition rates and thicknesses were measured using a quartz crystal thickness monitor. Thicknesses were determined following growth using variable-angle spectroscopic ellipsometry.

To obtain information about the composition of the degradation products, we measured the Fourier transform infrared absorption spectra (Nicolet 6700 FTIR spectrometer) from 100 cm<sup>-1</sup> to 4000 cm<sup>-1</sup> for 200 nm thick films deposited on CaF<sub>2</sub> windows (ESCO Products). The windows were cleaned using a dilute detergent solution, rinsed with deionized water, dried under N<sub>2</sub>, and further cleaned for 4 min with air plasma (Plasma Etch, PE-50). The films were aged for 1 week at ambient temperature. The FTIR sample stage was purged with N<sub>2</sub> prior to and during data collection, and the spectra were corrected for atmospheric water interference and baseline anomalies. Each spectrum was collected using 32 scans with a resolution of 4 cm<sup>-1</sup>.

A subset of deposited films was encapsulated with a glass lid sealed to the substrate using epoxy in an ultrahigh-purity N<sub>2</sub>

(< 1 ppm O<sub>2</sub> and H<sub>2</sub>O) environment. Aging was done in the dark or at  $35 \pm 5$  °C using a simulated AM1.5G illumination source at 1 sun intensity. Optical intensity was calibrated using an NREL-traceable Si reference cell. Absorbance was recorded by a Lambda 1500 spectrophotometer (Perkin Elmer). X-ray diffraction and atomic force microscopy measurements on as-grown and aged films were obtained in air.

## 3. Results

The absorption spectra of SubPc (11 nm thick) samples aged in air are shown in [Fig. 1](#). For the SubPc kept in the dark, there is a moderate decrease of the peak absorption at a wavelength of  $\lambda = 590$  nm after 30 h. However, when the SubPc sample exposed to 1 sun intensity, AM1.5G illumination, the same absorption peak disappears after 30 h, indicating complete bleaching of the SubPc film. In contrast, a SubPc:C<sub>60</sub> (1:4, 55 nm thick) blend shows no degradation when kept in the dark, and the decrease in absorption for a similar sample under illumination indicates that the rate of photobleaching is significantly slower, with only ~60% loss of the absorption peak at  $\lambda = 590$  nm after 67 h.

We also measured the time evolution of N<sub>2</sub>-packaged SubPc and SubPc:C<sub>60</sub> (1:4) film absorption as shown in [Fig. 2](#). After 2000 h, the reduction in SubPc film absorption is only ~10%, which is considerably less than the sample exposed to air ([Fig. 2a](#)). However, after 660 h, we observe a red shift of SubPc absorption at  $\lambda = 590$  nm, while the intensity of the shoulder at  $\lambda = 540$  nm increases slightly along with a similar wavelength shift. Similar trends are observed for the SubPc film aged under 1 sun illumination. After mixing SubPc with C<sub>60</sub> (1:4, SubPc:C<sub>60</sub>), the absorption

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