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Understanding coupled electro-thermal processes in the catastrophic failure of organic electronic devices

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ABSTRACT

The large-scale application of semiconducting polymers in organic electronics is hindered significantly by catastrophic device failure. Some of these processes are associated with coupled phenomena of electro-migration and thermal localization, which are poorly understood till date. In this paper we identify structural instability and its sources in a simple device configuration consisting of a semiconducting polymer sandwiched between two metal electrodes and subjected to various DC operating voltages. The coupled effect of temperature and electric fields leads to fracture of the polymer layer and the metal electrode layer, interestingly, in mutually exclusive regions. This failure is significantly observable beyond a certain applied voltage. It is observed that defects nucleate in a chain-like pattern with alternating fracture sites of polymer and metal electrode respectively. We subsequently propose a coupled electro-thermal mechanism which explains the observed phenomena. The mechanism is further validated by an analytical model of stress due to thermal and electric field distributions. The model predicts criteria of failure, which are interestingly complementary for the polymer and the electrode films, and hence explains the observation of the chain-like nucleation pattern. The failure criteria are functions of device geometry, operating voltage and temperature. This study will be useful toward reliability-based design of organic electronic devices including important factors such as coupled electro-thermal response and length-scale. The study also opens up important fundamental questions relating to the spatio-temporal evolution of electro-thermally induced minuscule electrical shorts in thin-film electronic devices.

1. Introduction

Organic electronics show promise for flexible thin film devices for display, sensing and energy harvesting applications including organic photo-voltaics (OPVs). Despite setbacks in performance compared to inorganic semiconductor devices due to material properties such as low charge carrier mobility, the possibility of inexpensive fabrication and to custom-design organic molecules to suit specific needs have favored extensive research in this area. However organic electronic devices, in particular, polymer electronic devices suffer a host of reliability issues which include problems in quantifying degradation of the material due to ambient exposure [1] and device-integrity issues which limit the operational life time. Of specific interest are thermal issues that limit the scalability of these devices to large areas [2–5]. It has been shown that under natural conditions, the temperature rise in large panels is significantly more compared to a single cell in isolation (Fig. 1) for various panel lengths L_p, obtained with the help of an empirical thermal transport model [6]. Organic light emitting diodes (OLEDs) operate at higher power densities of up to 3500 W/m^2 for practical Luminance of ~1000 cd/m^2 [7–11], which causes significant heating in a large panel. OPVs operate at lower power densities [12,13]. However excessive irradiance can cause heating. Additional sources of heat include the electrical circuits in panels and thermal absorption of the device package due to external heat, in which cases, heating scales up with panel sizes [14]. Partial shading and mismatch in radiative properties can cause excessive localized

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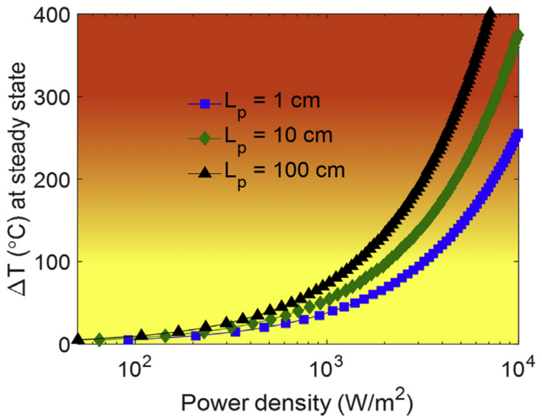

Fig. 1. Steady state temperature rise in flat panels as a function of operational power density, shown for various panel lengths L_p, predicted using empirical model (see Ref. [2]).

heating in panels [15]. Organic photodiodes are another class of devices which operate in a wide range of reverse bias voltages varying from 0 V to ~10 V [16]. However, their power densities are application specific and can lead to thermal problems. Thus, thermally induced failure phenomena are general to most organic electronic devices. Studies reported in the past include bubble formation in polymer LEDs caused by gas emission [17–19], accelerated growth of localized electrical shorts [20] and dark spots [21], although the mechanism of short formation is not clear. Other important factors are high electric fields and current densities leading to electro-migration of electrodes in polymer based OLEDs [22], diffusion of electrodes into the active layer [23,24], inter-diffusion of organic layers [25], failure due to thermal instability of organic layers [26,27], local electrochemical reactions [28], and delamination of active layers from electrodes [29].

While the above studies are indicative of the underlying problem, the coupled thermal and electrical phenomena are poorly understood, and are still being investigated [3,4,30,31]. On the fundamental front, studies on the response of dielectric polymers to simultaneous high temperatures and high electric fields have revealed the formation of instability driven structure [32]. However, it remains of great interest to study the coupled electro-thermal phenomena in organic electronic devices and we focus our effort here on this aspect. From a fundamental perspective, such studies would throw light on new mechanisms of electro-thermal behavior of polymers in contact with metal electrodes and help understand the mechanisms of formation of localized electrical shorts, and thermal hot-spots. From an applied perspective, it is of interest to study the effect of these mechanisms on the failure and life-time of the devices in order to create better design. Furthermore, such localized thermal and electrical effects are of direct relevance to geometrically induced effects in nanostructured optoelectronic devices, for example due to nano-structured electrodes [33–36]. The variation of inter-electrode distance in the nano-scale could lead to higher temperatures and electric fields, which could possibly lead to device failure. Here we study a thermally initiated structural degradation occurring in large area polymer diodes (~1 cm^2), with typical polymer layer thicknesses of around 100 nm. We identify and characterize a failure mechanism initiated by localized electrical heating in a device configuration consisting of a semiconducting polymer sandwiched between two metal electrodes. A certain regime of coupled localized electrical-heating and electric fields results in nucleation and growth of localized electrical micro-shorts in a chain-like nucleation pattern. This progressively reduces the device area and ultimately leads to catastrophic device failure. It leads to fracture of the polymer active layer and the metal electrode in mutually exclusive regions. Based on the experimental observations, a mechanism is proposed to explain the degradation phenomenon. To validate the hypothesis we derive an analytical model. Based on this model, we predict the stages of failure in terms of device geometrical parameters, temperature and applied voltage.

2. Details of experiment

The schematic top, cross-sectional, and isometric views of the device architecture are shown in Fig. 2(a). The device configuration consists of a semiconducting polymer Poly(3-hexylthiophene-2,5-diyl) or P3HT sandwiched between two metallic electrodes. The device parameters are listed in Table 1. Device fabrication details and measurements of active polymer film thicknesses are given in the supplementary section (SS-1 and SS-2 respectively). The ITO electrode is connected to the positive terminal of a voltage source and the Al electrode of region D1 is connected to the negative terminal. We increase the voltage gradually until visible deformations appear in the region, which are inferred by in-situ visual inspection, without the use of an optical microscope. At this point, the experiment is stopped and the structural deformations in the device are characterized by various techniques.

3. Experimental observation and discussion

First, we discuss the behavior of devices S1 and S2. As the bias voltage is increased to ~20V, visible deformations are observed in the bottom edge of the electrode of D1 (the region highlighted in Fig. 2(a)-top view), at the edge $y = 0$. This deformation is seen to propagate along $y > 0$ (see supplementary video and section SS-3 in supplementary information). The temperature of the device is around 130 °C (measured with an IR thermometer over a spatial range of ~1 mm) when the deformations become visible. Optical micrograph of the deformed region (Fig. 2(b)) shows that the deformations consist of a fracture pattern which fades away from the edge of D1 along $y > 0$. Also the representative fracture profile obtained by optical profiling (Fig. 2(d)), consists of two distinct fracture patterns at depths of 100 nm and 250 nm, below the top metal electrode. These depths closely match with the thicknesses of the Al cathode and the polymer film, respectively, in S2 on which the scan was performed, implying fracture of the electrode and possible defects in the active layer. . It is also seen that these features are not seen on a sample prior to stressing. Specifically, from Fig. 2(c), we see that the profile obtained on the device edge, on the top Al electrode, is significantly smooth compared to the deformed pattern seen in Fig. 2(d), after device stressing. An optical micrograph of a degradation site captured during its growth stage shows that the defect nucleates in chain-like fashion consisting of alternating dark and bright sites (inset of Fig. 2(b)). We call the dark and the bright sites as the ‘hinge’ and the ‘link’ sites, respectively.

Supplementary video related to this article can be found at http://dx.doi.org/10.1016/j.orgel.2016.10.011.

A comparison of changes shown in Fig. 3(a) and (c), indicates that the passive region D2 develops only a wrinkling pattern whereas the active region D1 shows fracture. However, the wrinkled morphology is common to both D1 and D2, although in D1 it is confined only to the vicinity of the edge $y = 0$.

The hinge sites are approximated by circular patterns of diameter of a few micrometers (Fig. 3(b) and (c)). EDS mapping is carried out on the region as shown in Fig. 3(c) and the spatial variation in the intensity for the elements Al, C and S are shown in Fig. 3(c1)–(c3). The elemental analysis of the hinge site from EDS