

Mapping the built-in electric field in polymer light-emitting electrochemical cells

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ABSTRACT

A millimeter planar polymer light-emitting electrochemical cell was turned on in a cryogenic probe station and subsequently cooled to freeze the doping profile. A 442 nm laser beam guided by an optical fiber was scanned across the interelectrode gap of several millimeters and the photovoltaic response was measured as a function of position. Both photocurrent and photovoltage profiles display a prominent peak at the geometric boundary of the p- and n-doped regions. A non-zero photovoltaic response throughout the p- and n-doped regions can be explained by various broadening mechanisms including non-uniform doping and secondary excitation by waveguided light. The photovoltaic response is weakest at the electrode/polymer interfaces.

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1. Introduction

Polymer light-emitting electrochemical cells (LECs) have been the subject of numerous recent studies all aimed at understanding the complex operating mechanisms of these devices [1–5]. LECs are unique among organic devices in that their active layer is a mixed ionic–electronic conductor consisting of a luminescent conjugated polymer and a solid-state polymer electrolyte [6]. The luminescent polymer is electrochemically p- and n-doped *in situ* via the application of a voltage bias. A light-emitting p–n junction is formed when the doped regions expand and make contact. The dynamic LEC doping process has been visualized by time-lapse fluorescence imaging of planar cells with an extremely large interelectrode gap [7,8]. The electronic structure of the LEC p–n junction, however, has not yet been adequately characterized.

LECs in a planar configuration have an exposed and highly scalable interelectrode gap that can be easily accessed both optically and electrically. Pingree et al. applied

scanning Kelvin probe microscopy (SKPM) to measure the potential profiles of μm -gap planar LECs under bias [9]. By varying the electrolyte salt and electrode materials, the group was able to shift the junction position and observe potential profiles well correlated with the optically imaged junction position [10]. The SKPM study by Matyba et al. on 120 μm planar LECs shows that the steepest potential drop occurs about 35 μm away from the cathode interface which is again consistent with the junction position [11]. Our group pioneered the turn-on and fluorescence imaging of planar LECs with millimeter interelectrode spacing [12]. The extremely large planar cells offer ease of fabrication and allow flexible testing schemes. For potential mapping, a 10.4 mm-planar LEC was turned on in a cryogenic probe station and promptly cooled to 200 K to freeze the doping profile. The potential drop due to an applied constant current was measured using a micro-manipulated tungsten probe in direct contact with the polymer film. The results establish that the doping profile of the planar LEC is that of a graded p–n junction. The doped polymer film is highly conductive and the p-doped region is more conductive than the n-doped region [13].

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Since an LEC once turned on is a p–n junction, it also possesses a built-in potential due to the chemical potential difference of the p- and n-doped polymers. Large, electrode-independent LEC built-in potential manifested as photovoltage has been observed in both sandwich and planar frozen-junction LECs under illumination [14,15]. LEC built-in potential is also responsible for the large open-circuit voltage observed when a fully charged sandwich cell is allowed to relax at room temperature [16]. In a planar cell configuration, the built-in potential could be spatially resolved using SKPM scans to show that it had indeed originated from the relaxing p–n junction [11]. For stable doping profiles, the optical-beam-induced-current (OBIC) technique is widely used to map the built-in potential or built-in electric field [17,18]. In OBIC measurements a focused light beam is scanned across the device and the photocurrent is detected whose magnitude is proportional to the strength of the local built-in electric field. The OBIC technique has been used to map the built-in potential of a 20 μm -planar LEC with a focused Ar ion laser beam [19]. Open-circuit voltage was detected in a narrow region ($\sim 2 \mu\text{m}$ wide) that was identified as the location of the p–n junction. Additionally, the photovoltage increased with the voltage bias that was applied to turn on the device. These results are consistent with the LEC operating mechanism by electrochemical doping and p–n junction formation. However, this initial LEC OBIC study preceded the discovery of frozen-junction LECs. The planar LEC was only cooled to 250 K after turn-on, which is above the glass

transition temperature of polyethylene oxide (PEO)–lithium triflate electrolyte. We have shown that at 250 K the junction is not sufficiently frozen [20]. This and the low excitation power used (nW) are likely responsible for the extremely weak (microvolt) photovoltage detected. Photocurrent was not measured to avoid rapid junction relaxation under short-circuit conditions. Here we present OBIC study of mm-gap planar LECs frozen at 200 K. Both photovoltage and photocurrent scans have been carried out along with photoluminescence and electroluminescence imaging of the device.

The planar LECs used in this study contained MEH-PPV, PEO (molecular weight 100 k) and cesium perchlorate (CsClO_4) in a weight ratio of 10:10:3. PEO and CsClO_4 were purchased from Aldrich and used as received. The LEC film was spin cast from a cyclohexanone solution and had thickness of $335 \pm 6 \text{ nm}$, as determined with an optical profiler. Co-planar gold and aluminum electrodes were evaporated on top of the LEC film using two sets of shadow masks in two steps. This involved the breaking of vacuum to switch the shadow masks for the second evaporation. The gold strip electrode has variable width so that each LEC has two interelectrode gaps of 3.1 mm and 4.6 mm, respectively. The cell was mounted in a Janis ST-500-1 micro-manipulated cryogenic probe station for testing under vacuum ($\sim 10^{-5}$ torr). Electrical contact to the device was made via two tungsten probes maneuvered by independent X–Y–Z translation stages. Tiny indium balls were attached to the tips of the probe to ensure good electrical contact to the

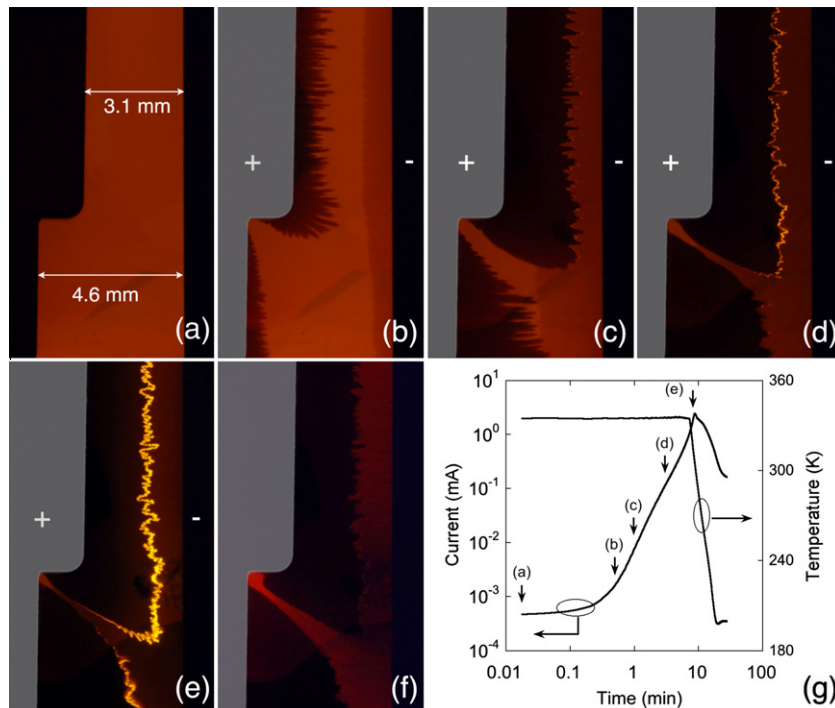


Fig. 1. Time-lapse fluorescence imaging of a MEH-PPV:PEO:CsClO₄ planar LEC with variable interelectrode spacing. The device was under 365 nm UV illumination and initially held at 335 K. The images were taken (a) 0 min, (b) 0.5 min, (c) 1 min and (d) 3 min after a 300 V bias was applied. Image (e) was taken at $t = 8$ min during cool down and at approximately 320 K. Image (f) shows the stabilized doping profile at 200 K without bias. (g) The cell current and temperature as a function of time during the turn on/cool down process. In (b–f) the gold electrode is highlighted for visibility. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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