Polymer Light-Emitting Electrochemical Cells-Recent Advances and Future Trends

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Abstract

Polymer light-emitting electrochemical cells (PLECs) are electroluminescent devices whose operation involves both ionic and electronic charges. A key reaction in PLEC is the electrochemical doping of the light emitter, a luminescent polymer, which eventually leads to the formation of a light emitting p-n or p-i-n junction in the interior of the cell. This review opens with a general introduction to the operating mechanism of PLECs. This is followed by a summary of key advancements in the field in the last two years. The focus of the review is on the chemical mapping and optical probing of the PLEC doping process and junction structures. The results showcased in this review provide a coherent picture of the PLEC operating mechanism and point in new research directions in both device applications and fundamental sciences of mixed ionic/electronic polymer conductors.

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I. Introduction

A light-emitting electrochemical cell (LEC) is a molecular electronic device developed alongside polymer and organic light-emitting diodes (PLEDs and OLEDs) for potential lighting and display applications. The term “electrochemical” implies the presence of ionic species by design. And electrochemical redox reactions involving both mobile ions and a light emitter are responsible for light emission from the cell. The light emitter in an LEC is invariably immobile and in direct contact with both electrodes. As such, LECs are solid-state electroluminescent (EL) devices and distinguished from liquid electrochemical cells wherein the light generated is typically electrochemiluminescent (ECL) in nature.[1,2]

The first LECs were polymer LECs or PLECs.[3,4] The active layer of a PLEC is typically a blend of a luminescent conjugated polymer as the electronic conductor and a polymer electrolyte as the ion conductor. LECs can be made with materials other than polymers.[5,6] This short review, however, concerns only PLECs and their key developments in the past two years. In particular, the focus is on the electrochemical and electronic structure of PLECs studied via various optical imaging and probing techniques. For information on other aspects of PLECs and other types of LECs, readers are urged to refer to recent reviews on the subject.[5,7,8]

Figure 1. Schematic illustration of PLEC device structure and operation. (a) Pristine PLEC without any voltage bias; mobile ions are uniformly distributed throughout the active layer. (b) PLEC under an applied voltage bias; *in situ* electrochemical doping first takes place at the anode and cathode interfaces. (c) The expansion of the p- and n-doped regions and the propagation of the doping fronts. (d) The formation of a light-emitting p-n junction; the PLEC is functional as a light-emitting device. (e) The schematic of a sandwich PLEC.

The presence of mobile ions (Figure 1 (a)) plays a central role in the PLEC operation. Upon the application of a sufficient DC voltage bias, injection of electrons and holes occur at the negative and positive electrodes, respectively, causing electrochemical doping of the luminescent polymer (Figure 1 (b)). In PLECs, doping refers to the insertion of counterions, such as $CF_3SO_3^-$ and K^+ , in the vicinity of an injected charge, which causes the semiconducting polymer to be doped, thus exhibiting vastly improved conductivity. The doping of the luminescent polymer poly (phenylene vinylene), PPV, is depicted by the following scheme.

Once the doping is initiated, the doping fronts move away from the electrode interfaces, leaving behind a doped region, as shown in Figure 1 (c). The presence of a doping front is necessitated by the higher conductivity of the doped region and can be numerically modeled.[9-11] The p- and n-doped regions continue to expand under the applied voltage bias until they meet to form a p-n or p-i-n junction, as shown in Figure 1 (d). This opens up a pathway for a significant electronic current. Junction EL results when injected electrons and holes recombine radiatively in the junction region.

The PLEC operation mechanism described above, also known as the electrochemical doping (ECD) model, was proposed by the original authors of the work to account for the unique device characteristics of PLECs.[3,12] Vastly reduced bulk and contact resistance, brought on by the high ion concentration and subsequent *in situ* electrochemical doping, readily explains PLEC's insensitivity to electrode work function and active layer thickness.[13] PLECs can therefore be made with an air-stable cathode and a thicker, more robust active layer. These traits make PLECs an appealing alternative to PLEDs. The PLEC's insensitivity to electrode work function and active layer thickness also makes it possible to fabricate functional planar cells, where the two electrodes (often the same metal) contact the same top or bottom surface of the active layer, with

some separation. The imaging of the original planar PLEC, fabricated on top of a pair of interdigitated gold electrodes with a gap size of 15 μm , offers the first visual evidence of a light-emitting p-n junction in an activated PLEC.[3] Later on, planar PLECs with a gap size of millimeters was demonstrated.[14,15] Extremely large planar PLECs have become an invaluable tool for the study of fundamental PLEC processes.[16-20] In Sections IV and V, time-lapse fluorescence imaging of large planar PLECs offer visual confirmation of the ECD model illustrated in Figure 1.

II. New materials and performance records in PLECs

In the past two years, PLEC research has advanced on many fronts. Device performance parameters such as peak luminance, efficiency and lifetime have all improved through better device and material engineering. By choosing the right barrier material, for example, an encapsulated PLEC built on a flexible substrate has been shown to be functional after a year in storage under ambient conditions.[21] PLECs have also been tested either continuously or intermittently, to extract an operational lifetime that can exceed 1,000 hours.[22-24] High performance electrolytes, based on star-branched oligomers, have been synthesized and applied to PLECs, leading to a record-high power conversion efficiency of 18.1 lm/W .[24] Figure 2 shows the turn-on kinetics and long-term stability of sandwich PLECs made with these high-performance electrolytes.

Figure 2. (a) Turn-on kinetics during the first min of operation and (b) the long-term stability of ITO/{SY + ion-transporter + LiCF₃SO₃}/Al sandwich-cell LECs, with the device structure disclosed in the inset of (a). The devices were driven by $j = 7.7 \text{ mA cm}^{-2}$. (c) The long-term stability of a TMPE-OC device driven by a prebias of 7.7 mA cm^{-2} for 40 min and thereafter by 1.9 mA cm^{-2} . (d) The temporal evolution of the efficiency for a TMPE-OC device equipped with a light-outcoupling film. The device was driven by a prebias of 7.7 mA cm^{-2} for 40 min and thereafter by 0.77 mA cm^{-2} . Reprinted with permission from (Mindemark, M.; Tang, S.; Wang, J.; Kalhovirta, N.; Brandell, D. and Edman, L., High-Performance Light-Emitting Electrochemical Cells by Electrolyte Design. *Chemistry of Materials* **2016**, 28 (8), 2618-2623). Copyright (2016) American Chemical Society.

The study of PLECs is in a large part motivated by their simple device structures and their potential as a low-cost alternative to existing or other emerging lighting technologies. Indeed, single-layer blue, white and near IR PLECs have been demonstrated with new emitters that are either donor- π -acceptor copolymers or donor/acceptor blends.[25-29] PLECs can also be driven from AC mains via an organic power converter and compatible with low-cost processing techniques.[30,31] The versatility of PLECs is further evidenced by the demonstration of stretchable, dual mode (electrochromic/emitting), biodegradable, or electrolyte layer mediated devices.[32-38] With a pulsed driving scheme, a peak current density in excess of $2,000 \text{ A/cm}^2$ has been realized in a planar PLEC. This record high current density, along with a direct doping charge measurement, again attest to the highly doped nature of functioning PLECs.[39,40]

III. Time-resolved chemical mapping in planar PLECs

The inherent complexity of a PLEC operation, which involves both ionic and electronic charges, has also produced fertile ground for the study of fundamental PLEC processes. The effect of salt anion on the performance of sandwich PLECs has been investigated.[41] The change in the refractive index of a LEC polymer has been measured as the polymer underwent controlled electrochemical doping. The results shed light on why the p- and n-doped polymer has different optical properties. [42] According to the ECD model, anions are mainly found in the p-doped region serving as counter ions for the oxidized luminescent polymer. This has been confirmed by a FTIR spectroscopic imaging study conducted on a planar PLEC.[43] Figure 3 shows side-by-side comparisons of the spectra and chemical intensity profiles of the PLEC before and after a 4 V bias has been applied. The triflate anions are traced by the SO_3^- vibration bands, and the oxidized MEH-PPV is identified by the C-C and ring stretching bands. The profiles clearly show that the anions are homogeneously distributed when the PLEC is in its pristine state. After being biased at 4 V for an hour however, a much higher density of anions is found on the anode side where the luminescent polymer MEH-PPV has been oxidized. This

study provides the first evidence of large-scale ion motion in a PLEC and complements the early profiling of sandwich PLECs by secondary ion mass spectrometry.[44,45] Time-resolved intensity mapping provides further support to the ECD model of PLEC operation.

Figure 3. FTIR line-averaged spectra of (a) the pristine sample and (c) the sample under 4 V applied bias after 1 h, obtained from lines I, II, and III in the reflection image (b) of the LEC device. The chemical intensity maps (d–k) were obtained over the same field of view as panel b and show integrated intensities over specific bands. For each pair of images, the left was obtained under zero bias (pristine sample), and the right was obtained under an applied bias. (d and e) Asymmetric and (f and g) symmetric SO_3^- vibration of the triflate anion; (h and i) C–C stretching and (j and k) ring stretching of MEH-PPV. The bands are as follows: asymmetric SO_3^- , 1290–1218 cm^{-1} ; symmetric SO_3^- , 1055–1007 cm^{-1} ; C–C stretch, 1441–1406 cm^{-1} ; ring vibration, 1503–1475 cm^{-1} . The field of view is $300 \times 300 \mu\text{m}^2$ in all images. Reprinted (adapted) with permission from (Jafari, M. J.; Liu, J.; Engquist, I.; Ederth, T., Time-Resolved Chemical Mapping in Light-Emitting Electrochemical Cells. *Acs Applied Materials & Interfaces* **2017**, 9 (3), 2747-2757). Copyright (2017) American Chemical Society.

IV. Scanning OBIC and PL imaging of a frozen PLEC junction

Since the PLEC is essentially a polymer p-n junction once activated, it is important to know the spatial extent of the junction depletion region. The optical beam induced current (OBIC) technique is well suited to extract this important junction parameter and has been applied to planar PLECs for this purpose.[46-49] In OBIC probing, a focused light beam is scanned across the planar cell and the resulting photocurrent is recorded. When photo-excitation occurs in the vicinity of the depletion region, a photocurrent is generated whose magnitude correlates with the local built-in electric field. In neutral p- or n-doped regions, by contrast, a null OBIC signal is expected due to the absence of any built-in field that can sweep the photogenerated charge carriers before they recombine. To perform an OBIC scan on a PLEC, a static doping profile and a straight junction are required to avoid any interference from the motion of ions or artificial broadening of the signal profile. In addition, the optical beam must be narrow and have a known profile. Third, the activated PLEC must have adequate conductivity to allow the minute photocurrent to be resolved against any background noise current. These conditions have been met in a recent study employing a focused laser beam and a large, frozen-junction planar PLEC.[50,51] Moreover, the scan can simultaneously record the photoluminescence (PL) intensity across the scan path.

Figure 4. Scanning OBIC and PL imaging of a frozen planar PLEC. (a) Time evolution of cell current and cell temperature during the activation process. A DC voltage bias of 20 V is applied. The inset shows the schematic of the planar PLEC. (b) Time-lapse fluorescence images of the planar LEC during the activation process. Only a section of the entire cell is shown. (c) Electrode-to-electrode OBIC and PL

scans. The cathode is located at $-230\text{ }\mu\text{m}$ and the anode is located at $470\text{ }\mu\text{m}$. (d) OBIC scan data of four scans along the same path and a Gaussian fit to the data near the OBIC peak. Reprinted (adapted) with permission from (AlTal, F. and Gao, J., High resolution scanning optical imaging of a frozen polymer p-n junction. *Journal of Applied Physics* **2016**, 120 (11), 115501). Copyright (2016) American Institute of Physics.

Figure 4 (a) shows the schematic of the $700\text{ }\mu\text{m}$ planar PLEC that has been activated and subsequently scanned. A 20 V constant bias voltage was applied to activate the PLEC, which was heated to 360 K. Under these conditions, the cell current rapidly increased to over 5 mA in about 50 s due to doping. The PLEC was subsequently cooled to 170 K to freeze the doping profile and the light-emitting junction formed. Figure 4 (b) shows the time-lapse fluorescence images of the PLEC before cooling. The orange PL of the light-emitting polymer MEH-PPV has been significantly quenched, revealing the doped regions. A light-emitting p-n junction is formed when the p- and n-doping fronts make contact. Figure 4 (c) shows the OBIC and PL profiles of a full (electrode-to-electrode) scan. The PL profile consists of an n-doped region to the left of higher overall PL and a darker, p-doped region to the right of the OBIC peak. A fairly broad ($\sim 30\text{ }\mu\text{m}$) transition zone is also detected between the two regions. The OBIC peak, by contrast, is extremely narrow. As expected, the OBIC signal is only detected in the junction region at the edge of the PL transition zone. Figure 4 (d) provides an expanded view of the OBIC profile that includes all data points from a total of four scans along the same path. The data are fitted to a Gaussian function with a $1/e^2$ width (defined as the width where the intensity has fallen to $1/e^2 = 0.135$ times the peak value) of $3.1\text{ }\mu\text{m}$, greater than the width of Gaussian excitation beam of $1.9\text{ }\mu\text{m}$. In all, an average junction width of $1.5\text{ }\mu\text{m}$ is determined from 18 scans along seven paths after deconvolution to remove the width of the excitation beam. These results illustrate the first time the depletion width of a frozen PLEC p-n junction has been resolved. The junction width is only 0.21% of the interelectrode gap, which is the lowest value measured among all PLECs.

V. PLECs with bipolar electrodes

Although called an electrochemical cell, PLECs are mainly studied as an alternative to PLEDs using the standard techniques and terminologies of organic and conventional semiconductor devices. Recently, the PLEC research has crossed into the field of bipolar electrochemistry, which has many applications but typically involves a liquid electrolyte.[52-56] Up to this point, it has been shown that the PLEC is a simple electrochemical cell terminated by two wired metal

electrodes, where the oxidation and reduction half reactions are initiated. The introduction of a floating conductor, in the interior of the PLEC, creates a *bipolar electrode*, or BPE that is capable of driving both reduction and oxidation reactions even though it is not directly connected to a power source.[57] This scenario is shown in Figure 5.

Figure 5. Time-lapse fluorescence images of a 1.94 mm planar cell consisting of one gold and one aluminum driving electrode, and a single aluminum disc BPE. The top part of the figure shows the cropped images of the cell around the BPE. The images were batch processed and uniformly enhanced in Photoshop. The cell was heated to 350 K and a 40 V bias was applied across the driving electrodes. The lower part of the graph shows the equivalent circuit of the cell when fully emitting. Reprinted (adapted) with permission from (Chen, S., Wantz, G., Bouffier, L., and Gao, J., Solid-state bipolar electrochemistry-polymer light-emitting electrochemical cells. *ChemElectroChem* **2016**, 3, 392-398). Copyright (2016) John Wiley and Sons Inc.

A single aluminum disc BPE of 0.26 mm in diameter is deposited on top of the active layer of a 1.94 mm planar PLEC. Electrochemical p- and n-doping, manifested as PL quenching, are not only induced by the driving electrodes, but also by the aluminum disc BPE. The doping reactions occur preferentially at the opposite extremities of the BPE where the potential difference

between the BPE and the PLEC film is the largest. The various doping fronts eventually meet to form four separate light-emitting p-n junctions, instead of the usual one. In PLECs with a linear array of BPEs, p- and n-doping originated from neighboring BPEs can interact to form multiple light-emitting p-n junctions in series.[58] With dispersed micro-BPEs, thousands of tiny light-emitting p-n junctions are formed within the bulk of the PLEC layer, creating a bulk homojunction PLEC.[59,60] A bulk homojunction PLEC, which is an analog of bulk ECL generation in a liquid cell,[61] exhibits vastly improved light output and giant open-circuit voltage when operated as a photovoltaic cell in the frozen-junction mode. BPE and bipolar electrochemistry, therefore, offer an elegant solution to the problem of narrow junction in PLECs. PLECs, in return, offer a brand new platform to conduct bipolar electrochemistry research in solid state. Solid-state bipolar electrochemistry complements conventional liquid electrolyte based bipolar electrochemical research and offers advantages such as simplicity and versatility in cell design and characterization. The solid state nature of the mixed ionic/electronic conductor, for example, allows for the demonstration of several new types of bipolar electrodes that are otherwise impossible to realize in a liquid cell. [62]

VI. Conclusions and outlook

PLECs are unique among organic photonic devices. The presence of mobile ions and the important role of doping and junction formation in PLEC operation distinguish PLECs from purely electronic PLEDs and OLEDs. PLECs exhibit highly desirable characteristics and have simple device structures. For practical applications, however, PLECs lag behind PLEDs and OLEDs in both operational lifetime and efficiency. The record lifetime and efficiency shown in Figure 2 has been achieved at relatively low luminance and current levels. At high luminance levels, PLECs suffer from severe efficiency roll-off due to increased level of doping that quenches the fluorescence of the polymer.[63] High intensity operation is also detrimental to the lifetime of PLECs.[22,64] An intermittent testing scheme, however, revealed that the fast luminance decay observed in PLEC stress test was in part caused by the reversible doping process and therefore largely recoverable.[65,66] This discovery suggests that the operational lifetime of PLECs could have been vastly underestimated. The ultimate solution to the fundamental tradeoff between efficiency and emission intensity [63] lies with frozen-junction LECs, in which all doping or side reactions involving mobile ions cease after junction formation.[67,68]. More research, however, is needed to realize frozen-junction LECs that have

sufficient shelf stability when stored at room temperature. The current record of about 1200 hours is realized by removing the ion-transporting polymer from the PLEC blends altogether. [69]. BPEs, especially their dispersed form, offer additional opportunities for efficiency enhancement by increasing the number of emitting junctions.[59,60] Fundamental research is also needed to better understand the complex doping processes and the electronic structures of the PLEC junction. In particular, we need to experimentally answer the question-how does the junction width scale with the separation between the driving electrodes or BPEs? Here, extremely large planar PLECs are a valuable and versatile tool due to easy access, via both electrical and optical probes, to the active layer. The study of PLECs, both as a viable light-emitting device as well as a visual model system could also be of potential interest to the wider electrochemical device community.

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