

## On the desired properties of a conjugated polymer-electrolyte blend in a light-emitting electrochemical cell

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### ABSTRACT

We present results from a systematic study on the influence of the conjugated polymer (CP) on the performance of planar light-emitting electrochemical cells (LECs) with a device structure of Au/{CP + poly(ethylene oxide) (PEO) + KCF<sub>3</sub>SO<sub>3</sub>}/Au. We have employed six different CPs, and we demonstrate that in order to attain a fast turn-on time and a strong light emission intensity, it is critical that the p-type doping (oxidation) potential of the CP is situated within the electrochemical stability window of the {PEO + KCF<sub>3</sub>SO<sub>3</sub>} electrolyte. We also find that a high ionic conductivity of the active material correlates with a small phase separation between the CP and the {PEO + KCF<sub>3</sub>SO<sub>3</sub>} electrolyte, and that a doping concentration of ~0.1 dopants/CP repeat unit is a generic feature of the progressing doping fronts in all investigated devices. Finally we report the first observation of a light emission zone positioned in close proximity to the positive anode in a CP-based LEC.

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## 1. Introduction

Light-emitting electrochemical cells (LECs) have attracted increasing interest recently, mainly because they offer efficient light emission at low voltage from potentially easy-to-fabricate devices, but also because their complex and fascinating turn-on process has stirred a lot of debate regarding even its fundamental nature. Two main types of LECs exist: small-molecule based LECs, [1–14] and conjugated-polymer based LECs [15–34]. The former are commonly based on an ionic transition metal complex as the single-component active material, while the latter typically contain a three-component active material mixture, comprising a conjugated polymer (CP), an ion-solvating and ion-transporting material, and an alkaline salt.

We have recently set out on a quest to investigate and establish the desired properties of the various components in CP-based LECs for the attainment of optimized operation. We have established and reported that the physical and electrochemical properties of the electrolyte (*i.e.*, the dissolved salt together with the ion-solvating and ion-transporting material) and the electrode material play a crucial role. For instance, we find that the size of the alkaline-salt cation directly correlates to the device turn-on time and the light emission intensity [35,36], and that the electrochemical stability window of the electrode material is relevant in the context of elimination of undesired electrochemical side reactions [37]. In this paper, we extend these studies to include the core component of such devices, *viz.*, the CP.

Our view of the *optimum* turn-on process of a CP-based LEC is as follows: when a voltage equal to or larger than the band-gap potential of the CP is applied between the two

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electrodes ( $V \geq E_g/e$ ), the dissolved ions redistribute in the resulting electric field to form electrical double layers (EDLs) at the electrode/active material interfaces [38]. The voltage drop over these (sub-nm-) thin EDLs is of such a magnitude that it compensates for the difference in energy between the work function of the electrode and the appropriate energy level of the CP, thus allowing for efficient oxidation of the CP (hole injection) at the anodic interface and efficient reduction of the CP (electron injection) at the cathodic interface. The injected electronic charge carriers are thereafter electrostatically compensated by the migratory motion of dissolved ions. The process of injection of holes/electrons onto the CP together with the subsequent electrostatic compensation of ions is termed electrochemical p-type/n-type doping.

At a certain doping concentration, the CP exhibits a relatively distinct transition from being an electronic insulator to become an electronic conductor, with the consequence that the metal electrodes now are in contact with a layer of highly conducting doped CP. Accordingly the electrochemical action (i.e., the injection of electronic charge carriers from an electronically conducting phase to an electronically insulating and ionically conducting phase) now takes place at the interface between the doped CP and the undoped CP (i.e., at the doping front) instead of at the metal interface. The p-type and n-type doping fronts continue to move towards each other in the inter-electrode gap, until they make contact to form a p–n junction. At this point the device has turned on, and subsequently injected electrons and holes can recombine under the emission of light at the p–n junction.

However, it is important to emphasize that the above outlined scenario represents the turn-on process of an ideal LEC, and that a number of non-desired effects can take place in a real device. For instance, it is possible that the balanced electrochemical reactions at the anodic and cathodic interfaces during device turn-on *not* correspond to p-type doping and n-type doping, respectively, of the CP, but instead involve an irreversible side reaction involving the electrode or the electrolyte material. Moreover, it

has been demonstrated in a number of publications that the CP and the electrolyte phase separate [39–44], and it is not completely clear how this phase separation influences the turn-on process of an LEC.

In this paper, we have employed planar LECs with extremely large mm-sized inter-electrode gaps to enable for a direct optical probing of the device turn-on process; see Fig. 1 for a schematic of the planar device structure. We have consistently employed Au as the electrode material and KCF<sub>3</sub>SO<sub>3</sub> dissolved in poly(ethylene oxide) (PEO) as the electrolyte, so that a direct correlation between device data and the chemical structure of the CP can be obtained. We have also characterized the active materials (i.e., the CP blended with the electrolyte) as regards to phase separation, electronic structure and electrochemical reversibility, and correlated these results with the acquired device data. We find that the ionic conductivity of the active material, as expected, benefits from a small phase separation between the CP and the electrolyte. More surprising, however, is that we find that an electrochemical side reaction involving the {PEO + KCF<sub>3</sub>SO<sub>3</sub>} electrolyte in several cases hinders, and in some cases even eliminates, the p-type doping process of the CP and the subsequent device turn on and light emission in LEC devices. We rationalize these findings by demonstrating that the p-type doping potential of the CP in these specific cases exceeds the oxidation stability potential of the electrolyte, which in turn provides an important guideline for the selection of appropriate CP/electrolyte combinations.

## 2. Experiment

### 2.1. Materials

MEH-PPV ( $M_w = 150,000$  g/mol; Aldrich) is commercially available and was used as received. The procedures for the synthesis of BEH-PPV, BUEH-co-MEH-PPV, BUEH-PPV, BOP-PPV and BEHP-co-MEH-PPV are available in the literature [45–48]. The chemical structures of the CPs are presented in Fig. 2. Poly(ethylene oxide) (PEO,  $M_w = 5 \times$

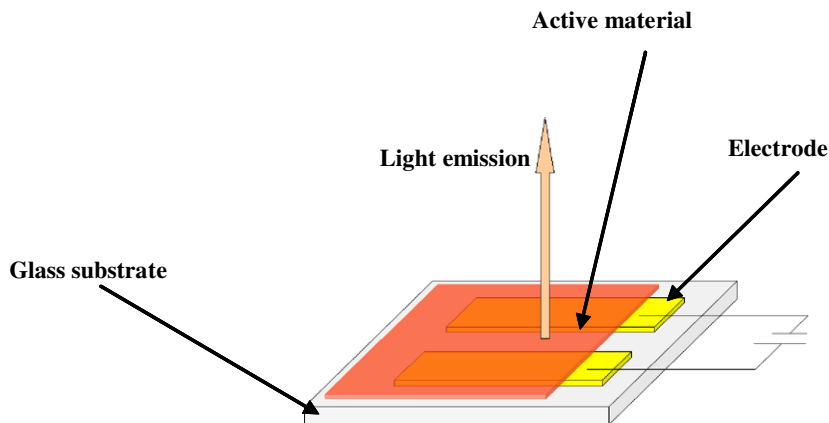


Fig. 1. Schematic of the employed planar LEC device structure deposited on a transparent glass substrate, with the electrodes positioned in a bottom-electrode configuration.

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