



Controlling the dynamic behavior of light emitting electrochemical cells



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ABSTRACT

Light emitting electrochemical cells (LECs) present an attractive route towards cost efficient lighting applications. By utilizing ionic phosphorescent transition metal complexes, efficient electroluminescence can be realized from a single layer device using air stable electrodes. These devices achieve efficient charge carrier injection due to ion accumulation at the interface upon driving, resulting in a dynamic response upon device operation. Here we investigate the device operation by using fast current and luminance versus voltage sweeps during normal fixed bias operating. A universal set of JL – V curves can be identified in which different regimes are observable. The speed and extent in which a LEC evolves through this set of curves can be controlled by varying the driving voltage, enabling the device to operate in its maximum efficacy state.

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1. Introduction

Organic light emitting diodes (OLEDs) are considered a viable candidate for large-area, low-cost lighting sources since their efficiencies and stabilities have improved significantly over the last years [1]. Most of these devices however require rigorous encapsulation in order to prevent degradation and are based on a multilayer architecture. Such multilayer devices are typically fabricated by vacuum evaporation of small molecular weight components. All these factors hinder the realization of a truly cost effective lighting solution. Light emitting electrochemical cells (LECs) provide an interesting way to overcome these complications by the incorporation of mobile ions in the active layer [2–4]. During operation, accumulation of these ions at the interface facilitates charge injection and enables efficient electroluminescence from single active layer architectures using air stable electrodes [5–7]. This allows for their preparation using solution-based techniques, which makes them suitable for low-cost and large-area applications. A wide range of emission colors, including white,

and efficiencies as high as 36 lm W^{-1} have been reached with iridium(III) ionic transition metal complexes (iTMC) [5,8]. Additionally, stabilities in excess of 4000 h at an average luminance of 650 cd m^{-2} have been reported for these type of electroluminescent devices [9].

Under the application of a bias, the current and luminance in LECs slowly increase in a timescale ranging from milliseconds to hours. This behavior has been generally described in terms of the essential property governing LEC operation, which is the separation of ionic charges [10,11]. Firstly, the application of a bias leads to ion accumulation at both interfaces resulting in the formation of large interfacial fields, reducing the barrier for carrier injection. A consequence of this electric double layer formation is a screening of the external electric field rendering the bulk of the device effectively field free [12]. At sufficiently high applied biases the existence of a field-free regime in the bulk disappears [13]. In this case injected carriers are electrostatically compensated by ions of the opposite sign diminishing the buildup of space charge [14]. Since the amount of dissociated ions is limited, compensation of electronic charges does not occur in the centre of the device resulting in a large potential drop in that region [15]. The combination of (relatively) immobile ionic

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species and electronic charge can be referred to as electrochemical doping [16], hence a *p*-doped/intrinsic/*n*-doped (*p-i-n*) structure is formed [17–19]. The exact bias and operation time at which the breakdown of the field free regime and formation of a *p-i-n* geometry occurs, depends strongly on the amount of ions in the film, carrier mobilities, injection barriers, film thickness and feedback mechanisms modifying the rate of carrier injection [20].

For the proper operation of LECs it is crucial to understand and control the dynamic response of LECs. Here we investigate the dynamic behavior of an archetype iTMC based LECs under the application of a fixed bias. Sandwiched LECs based on an ionic iridium complex emitting intense orange light were used for this study (Fig. 1). Due to the slow response of these devices of up to hundreds of hours, they provide an excellent opportunity for the separate monitoring of ionic and electronic properties of the devices. For this, at specific and very short time intervals during the fixed bias operation, fast current density (*J*) and luminance (*L*) versus voltage (*JL-V*) sweeps are performed. Care is taken to ensure that the ionic distribution is not disturbed during the sweeps such that they probe the electrical properties of the system. Previously, this technique has been shown to give valuable insights in the device operation of LECs operated at a fixed voltage of 3.5 V [15]. It was shown that there is a specific moment during the operation of a LEC at which the turn-on voltage for light emission has reached a minimum and the efficacy has become voltage independent, indicating that the device is no longer injection limited. With increasing operating time the current density increases even further and is shown to follow a space-charge limited behavior typical of normal OLEDs, characterized by a quadratic dependence on voltage and a third order dependence on the effective thickness. The increase in current originates from a decreasing thickness of the intrinsic zone which was demonstrated by impedance spectroscopy [15]. Due to the decreasing intrinsic zone, excitons generated in that zone are more prone to quenching by the radical cations and anions present in the doped layers, which leads to a decrease in the device efficacy [21].

Thus, for operating voltages above the bandgap of the emitting species and at long timescales electrochemical doping processes dominate the device characteristics lead-

ing to the undesired decreasing of the intrinsic region. The question remains, whether by using other operating voltages, one can control this behavior, and more importantly, if the continued shrinking of the intrinsic region and associated loss in efficiency can be avoided. To try and elucidate this question we performed and analyzed fast *JL-V* sweeps at set intervals on LECs that were operating under a constant bias of 1.8, 2.5, 3.5 and 4.5 V. For each of these devices the luminance, current density, efficiency and turn-on time are determined from the *JL-V* sweeps at the 3 V point. This allows us to directly compare these differently operated LECs, which would be impossible otherwise due to the interdependence of both the electronic and ionic currents on applied bias. The current density of the low voltage (1.8 V) operated LEC follows the trend predicted by the theory of double layer formation. Yet, by rapidly scanning to a voltage of 6 V we can obtain information on the turn-on voltage for light emission which significantly reduces from 5.5 to 4 V over the first 20 h. For the higher voltage operated LECs we show that the maximum efficiency is independent on the driving voltage and that it occurs at identical current density and luminance levels. This is new and striking information that confirms that the real parameter to control is the current density.

2. Materials and methods

The studies presented are performed on sandwiched LECs that consists of a layer of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS, CLEVIOS™ P VP Al 4083, aqueous dispersion, 1.3–1.7% solid content, Heraeus) (80 nm) on top of a patterned indium tin oxide (ITO)-coated glass substrate (<http://www.naranjosubstrates.com>) followed by the active layer that consists of bis(2-phenylpyridine-*C,N*)(2,2'-bipyridine-*N,N'*)iridium(III) hexafluorophosphate [Ir(ppy)₂(bpy)]⁺(PF₆)⁻ (90 nm), mixed with the ionic liquid 1-butyl-3-methylimidazolium hexafluorophosphate, (BMIM⁺)(PF₆)⁻ (>98.5%, Sigma-Aldrich) at a molar ratio of 4:1 spin-coated from an acetonitrile solution. The thickness of the films was determined using an Ambios XP1 profilometer. After spinning the organic layers, the samples were transferred to an inert atmosphere glovebox (<0.1 ppm O₂ and H₂O, MBraun) and dried on a hot plate at 80 °C for 1 h. Aluminum metal electrodes

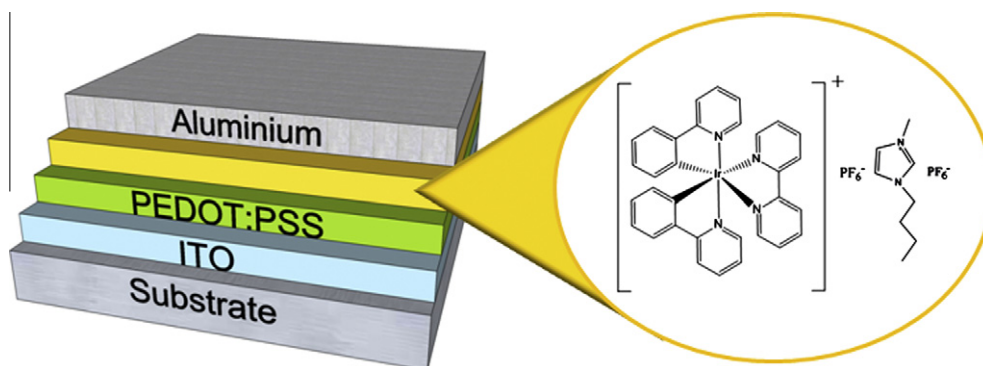


Fig. 1. Layout of the sandwiched LEC device and chemical structures of the iridium iTMC and the ionic liquid used in the active layer.

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