

Strategies toward long-life light-emitting electrochemical cells

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Abstract

This mini-review examines the operational lifetime of light-emitting electrochemical cells (LECs). Under continuous operation, both polymer-based LECs (PLECs) and ionic transition metal complex (iTMC)-based LECs (iTMC-LECs) now exhibit a luminance half-life exceeding 1,000 hours. This improved performance is accomplished with several effective strategies aimed at optimizing the operating scheme, the material composition, and the device architecture. These strategies are presented in detail with PLECs as an example. iTMC-LECs are also highlighted due to their excellent stress stability with regards to both luminance and operating voltage. The survey of literature data points to clear trends, as well as some unexpected results in LECs stressed for an extended period. Major challenges still exist, but long-lasting LECs are possible when the proven strategies are combined with innovative materials and device design.

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I. Introduction

Light-emitting electrochemical cells (LECs) are solid-state electroluminescent (EL) devices whose active medium is a luminescent, mixed ionic/electronic conductor.[1-3] The intentionally added mobile ions distinguish LECs from other types of molecular devices. First developed in the 90s as an alternative to the polymer light-emitting diodes (PLEDs), LECs now employ a variety of luminescent materials such as conjugated polymers (CPs),[4-11] organic small molecules (SMs),[12-14] and ionic transition metal complexes (iTMCs)[15-22]. LECs are attractive molecular devices for potential display and lighting applications due to their simple device structure, compatibility with low-cost solution processing techniques, and highly desirable device characteristics.[23,24]

Prototypical PLECs are based on a composite of a luminescent CP and a polymer electrolyte.[25,26] The latter is typically a complex of polyethylene oxide (PEO) and a lithium triflate salt (LiTf). The presence of mobile ions, contributed by the polymer electrolyte, causes *in situ* electrochemical p- and n-doping of the luminescent CP when they interact with injected holes and electrons. The doped CP exhibits vastly increased conductivity. As a result, LECs are insensitive to electrode work functions and active layer thickness. This affords LECs an efficient single layer construction, thicker active layers, and air-stable electrode materials. Light emission in a LEC takes place when the contrarily moving doping fronts make contact to form a p-n or p-i-n junction.

The introduction of ionic charges, on the other hand, also complicates the LEC processes. The LEC doping process and junction structure continue to be an important subject of research all on their own.[27-33] Over the past twenty years, our understanding of LECs has greatly improved. LEC device performance, however, is still lagging behind that of PLEDs or OLEDs in the operational lifetime. For example, state-of-the-art, solution-processed PLEDs created by Cambridge Display Technology (CDT) exhibit a T95 lifetime (accelerated tests) of 5,800 hrs and 7,500 hrs respectively for red and green-emitting devices with an initial luminance of 1,000 cd/m². The T50 lifetimes are on the order of hundreds of thousands of hours.[34] By comparison, the longest reported LEC lifetime is about two orders of magnitude lower. This review seeks to identify effective strategies that can address this key deficiency of LECs. PLECs will be used as a primary example and examined in detail. The review points to a challenging, yet also promising future for the realization of long-life LECs. The review is organized into nine sections.

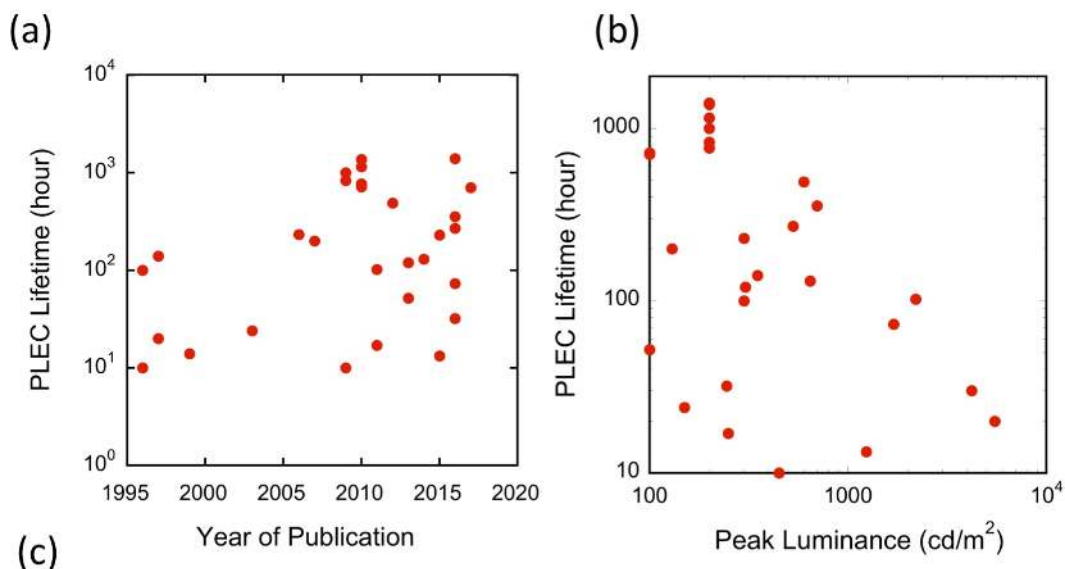
In Section II, a literature survey of PLEC lifetime data is given. In Sections III-VI, various strategies for long-life PLECs are discussed. In Section VII, long-life iTMC-LECs are introduced. Section VIII outlines the challenges and opportunities for an improved LEC operational lifetime. Finally, Section IX provides conclusions and an overall outlook.

II. Literature Survey of PLEC Lifetime Data

Figure 1 captures representative PLEC operational lifetime data reported over the past 20 years. The data are limited to studies with a sufficient test duration (≥ 10 hours) and/or peak luminance (100 cd/m^2). In Figure 1(a), the PLEC lifetime is plotted against the year of publication. In general, PLEC operational lifetime data are quite scarce when compared with other performance figures of merit such as peak luminance or efficiency. This is especially true in the first decade after the invention of PLECs. The reporting of such data, however, increased in volume around 2009, when an operational lifetime of $\sim 1,000$ hours was first noted. It should be cautioned here that the PLECs in these studies differ significantly in their structures, materials, and test conditions. And the operational lifetime, as a key figure of merit, was not defined in the same way in these studies. Lifetime values are directly quoted from the publications without any attempt to extrapolate, calibrate, or normalize. Some represent the time needed for luminance to drop below a certain percentage (50%-95%) of peak luminance,[8,35-45] while some are simply the test durations,[46-49] and some are defined as the duration wherein the luminance is above a certain level (100 cd/m^2 or 300 cd/m^2).[6,50-54]

Despite the lack of a standard definition, clear patterns emerge when the lifetime data are plotted against the peak luminance of tested cells, as shown in Figure 1(b). The longest lasting PLECs, concentrated in the top left, were all operated at a low peak luminance of $100\text{-}200 \text{ cd/m}^2$. [6,50,52] As expected, operational lifetime decreases with the peak luminance of the tests. On the bottom right of the graph, one study stands out for its high peak brightness up to $5,500 \text{ cd/m}^2$. [42] When operated at $2,200 \text{ cd/m}^2$, the cell exhibited a T75 lifetime of 102 hours. The extrapolated lifetime at 100 cd/m^2 reached a record 27,000 hours. The details of these longest-lasting cells are summarized in Figure 1 (c). Also included in Figure 1 (c) is a white PLEC with a very promising T80 lifetime of about 13 hours when tested with a high peak luminance of over $1,200 \text{ cd/m}^2$. Many considerations went into the design and testing of these high-performing PLECs. In the following sections, some of the design strategies will be examined regarding their rationale, merits, as well as drawbacks. In doing so, we hope to synthesize new routes that can

potentially lead to even longer lifetimes.



PLEC Structure	Operating Current	Peak Luminance	Lifetime (hour)	Peak Efficiency
ITO/PEDOT:PSS/MEH-PPV:PEO:KCF ₃ SO ₃ /Al	5 mA for 0.5 hour, followed by 1 mA	~200 cd/m ²	~1,000	1.9 lm/W
ITO/PEDOT:PSS/SY:TMP E:LiCF ₃ SO ₃ /Al	7.7 mA/cm ² prebias followed by 1.9 mA/cm ²	~200 cd/m ²	1,150	8.6 lm/W 8.6 cd/A
ITO/PEDOT:PSS/SY:PS:T MPE:LiCF ₃ SO ₃ /Al	7.7 mA/cm ² prebias followed by 1.9 mA/cm ²	~200 cd/m ²	1,375	10.2 lm/W
ITO/SY:TMPE-OC:LiCF ₃ SO ₃ /Al	7.7 mA/cm ² for 40 min followed by 1.9 mA/cm ²	~200 cd/m ²	1,400	18.1 lm/W
ITO/SY:PEO-DMA:LiCF ₃ SO ₃ /Al	25 mA/cm ²	2,200 cd/m ²	102 (T75)	15.3 lm/W
ITO/WLEP:ETPTA:LiCF ₃ S O ₃ /Al	13.3 mA/cm ²	1,241 cd/m ²	13.3 (T80)	8.1 cd/A

Figure 1. (a) Published PLEC operational lifetime vs. year of publication. (b) Published PLEC lifetime vs. peak luminance of stress test. (c) Details of some long-life PLECs from references [6,42,50,52,55]

III. Optimizing electrolyte loading

Early PLECs typically contained a large quantity of electrolyte materials. For example, a weight ratio of PPV:PEO: LiTf=50:50:8.9 was used in one of the original PLECs.[25] This corresponds to an EO/ Li⁺ molar ratio of 20:1, which has been shown to exhibit the highest ionic

conductivity in PEO:LiTf complexes when the salt concentration is varied.[56] The PLECs, containing more than 50% electrolyte by weight in their active layers, exhibited symmetric light intensity vs. voltage (L vs. V) and antisymmetric current vs. voltage (I vs. V) characteristics that are unique to LECs. PLECs with high electrolyte loading, however, exhibited a T50 lifetime of only 6-10 hours and were slow to turn on. This has been attributed to the poor film quality of the PLEC active layer, which was comprised of two incompatible high polymers.[35]

A 2009 study explored the effect of electrolyte loading on the operational lifetime of MEH-PPV:PEO:KCF₃SO₃ LECs.[50] Starting from an active layer composition of MEH PPV:PEO:KCF₃SO₃=1:1.35:0.25, which corresponded to a EO/K⁺ molar ratio of 23:1, the salt content was progressively reduced to 1:1.35:0.03. Reducing the salt concentration was found to incur a minimal loss of power efficiency or a loss of operational lifetime. Next, the PEO content was reduced with the MEH-PPV and salt contents fixed at 1:0.03. This, however, resulted in a 30-fold increase in the operational lifetime when the PEO content was lowered from 1:1.35 to 1:0.085, as shown in Figure 2 (a). A record lifetime of nearly 1,000 hrs had been achieved with an optimized electrolyte amount. This is shown in Figure 2 (b).

Because PLEDs based on an orange-emitting polymer similar to MEH-PPV exhibited a lifetime of over 10,000 hours,[57] it is highly probable that the poor lifetime of PLECs is caused by adverse reactions involving the electrolyte. Reducing the electrolyte loading, so long the doping process is not impeded, should benefit the stability of PLECs. The results of Figure 2 demonstrate the effectiveness of such an approach. In fact, all PLECs listed in Figure 1 (c) had a low electrolyte content of about 10%-20% by weight. These PLECs also exhibited high efficiencies and characteristic LEC turn-on/activation behaviors. A tradeoff of this approach is a slow turn-on response under a fixed voltage bias, as shown in Figure 2 (a). This drawback can be overcome by activating the PLEC with a variable current driving scheme, as shown in Figure 2 (b). Reducing the amount of electrolytes, however, does not solve the PLEC longevity problem in its entirety and is unsuited for application in extremely large planar PLECs. PLECs with reduced electrolyte loading also lose the symmetric/antisymmetric L-V/I-V characteristics. [58] The unique device behaviors of PLECs under reverse bias operation will be discussed in Section IV and Section VIII.

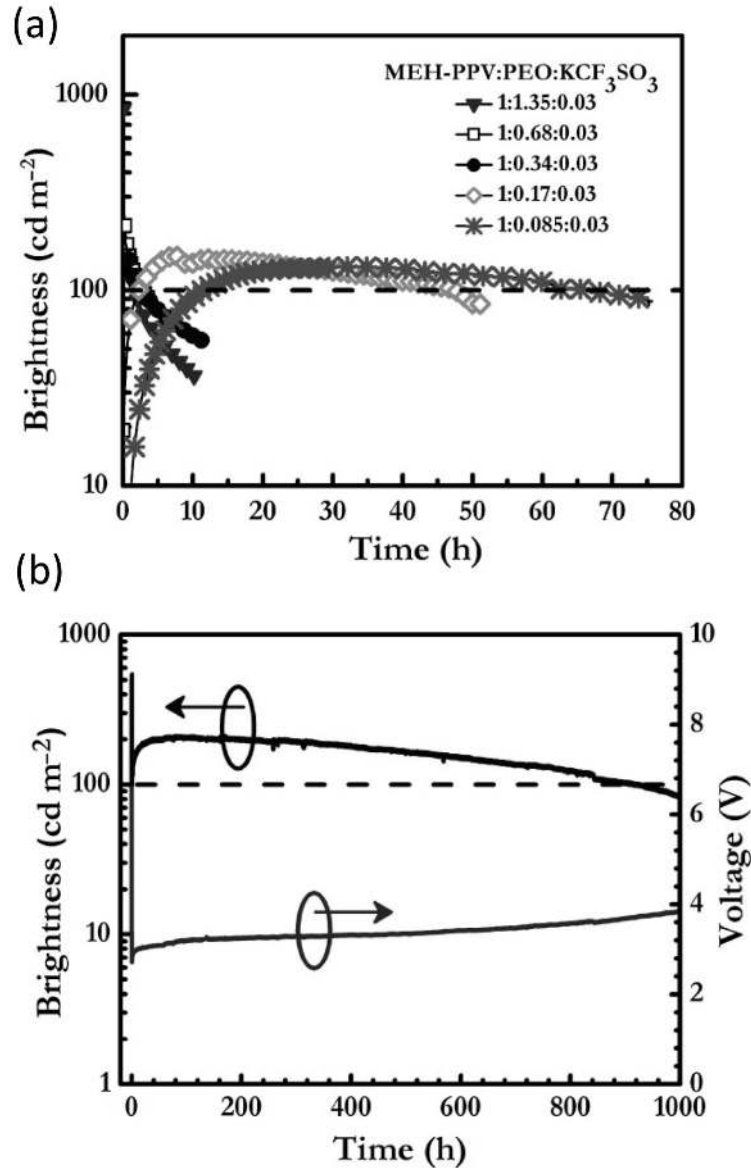


Figure 2. (a) The temporal evolution of the brightness of ITO/MEH-PPV:PEO:KCF₃SO₃/Al sandwich cells with different PEO concentration in the active material, as specified in the upper inset. The devices were tested at V=3 V and T=360 K. (b) The temporal evolution of the brightness (upper line) and the voltage (lower line) of an ITO/PEDOT-PSS/MEH PPV:PEO:KCF₃SO₃/Al sandwich cell with an active material mass ratio of 1:0.085:0.03. The device was operated at T=298 K and in galvanostatic mode. The initial “pre-bias” current, $I_{\text{pre-bias}}=0.005$ A, was applied for $t=0.5$ h, and it was followed by long-term operation at $I=0.001$ A. Reprinted (adapted) with permission from (Fang, J., Matyba, P., and Edman, L., The Design and Realization of Flexible, Long-Lived Light-Emitting Electrochemical Cells. *Advanced Functional Materials*, **2009**, 19, 2671-2676). Copyright (2009) WILEY-VCH.

IV. Freezing or chemically fixing the PLEC junction

To completely overcome the main drawbacks of PLECs, namely fast degradation and a slow turn-on response, it is desirable that a PLEC, once activated, de-couples itself from any ionic processes. In other words, a PLEC with a fixed junction, much like an inorganic p-n or p-i-n junction LED, is needed. The route to a fixed PLEC junction lays with the polymer electrolyte, which has been the subject of intense research effort due to its applications in rechargeable batteries, fuel cells, and recently, dye-sensitized solar cells.[59,60] Ion transport in a polymer electrolyte has been known to be highly dependent on temperature. In PEO/Li⁺ complexes, for example, cation transport is enabled by polymer chain segmental motion, which forms and breaks coordination bonds between the Li⁺ ion and the ether oxygen. The chain motion also creates “free volume” that provides space for ions to move into. The amorphous phase of the PEO/Li⁺ complex is mainly responsible for the ion conductivity, whose temperature dependence can be described by an exponential relation known as the Vogel-Tamman-Fulcher (VFT) equation.[61] Since polymer chain segmental motion ceases below the glass transition temperature (T_g), a freeze-out of ion motion and conduction also occurs. This important polymer electrolyte property was exploited to create a “frozen-junction” LEC in 1997.[62] A ITO/MEH-PPV:PEO:LiTf/Al PLEC was activated at room temperature and subsequently cooled to 100 K, which was well below the T_g of PEO:LiTf complex at about 208 K.

Frozen-junction PLECs exhibit sub-microsecond response times and diode-like I-V and L-V characteristics. Significant current flow and EL only occur when the bias polarity is the same as that of the activation voltage bias. More importantly, frozen-junction PLECs exhibit enhanced stability and minimal efficiency roll-off even when driven to a current density of 300 mA/cm². When driven with a constant current density of 5 mA/cm² at 200 K, EL intensity decreased only by 5% from the initial value after more than 10 hours of continuous operation.[38]

Fluorescence imaging of extremely large planar PLECs offer direct evidence of a static doping profile at temperatures below T_g .[63] A stress test of such frozen planar PLECs initially yielded puzzling behaviors--both driving voltage and EL intensity increased rapidly over an extended period of more than 150 hours.[64] This was later attributed to the effect of self-heating by the device being tested.[65] The heating caused partial relaxation of the frozen junction, which in turn made the device more resistive but less quenched, hence the observed increase in both EL intensity and driving voltage. The relationship between doping-induced quenching and EL intensity/decay will be discussed in more detail in Section V. By fabricating the planar

PLECs on a sapphire substrate instead of a glass substrate, the real cell temperature was close to that of the cold finger due to sapphire's excellent thermal conductivity. This resulted in a "normal" stress response in EL intensity and driving voltage, as shown in Figure 3.

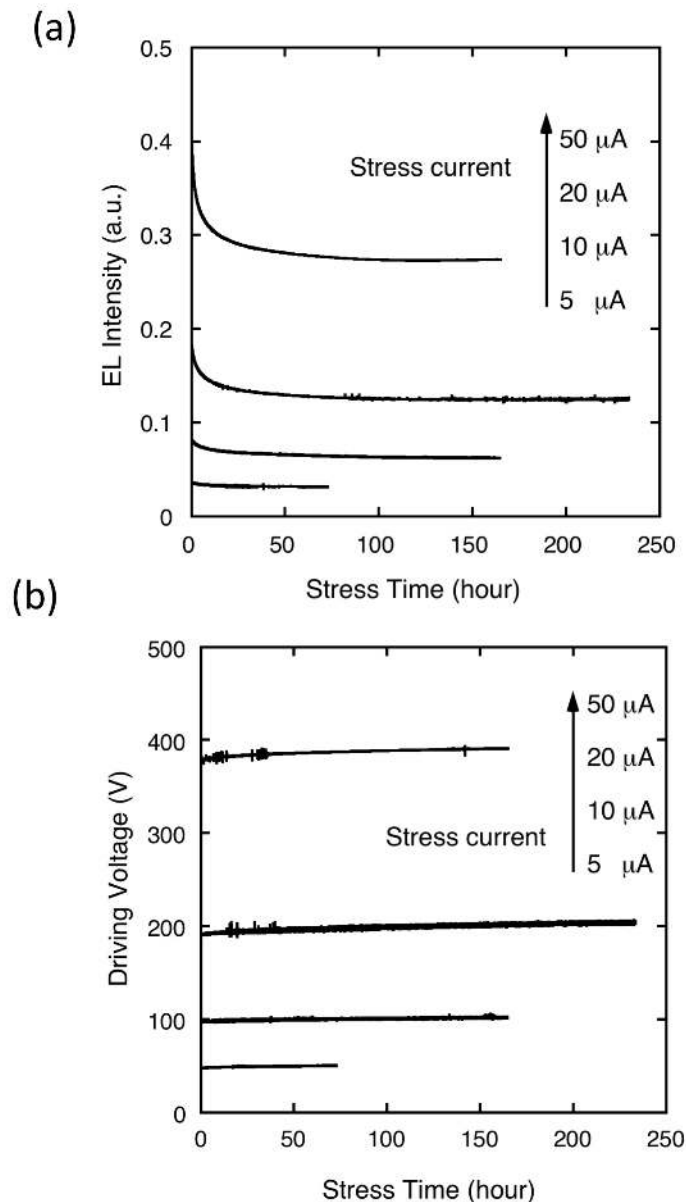


Figure 3. (a) Time evolution of EL intensity for identical 0.6 mm frozen-junction planar PLECs operated under various currents at 200K. (b) Time evolution of device driving voltage for cells operated in (a). Reprinted (adapted) with permission from (Zhang, Y. and Gao, J., Lifetime study of polymer light-emitting electrochemical cells. *Journal of Applied Physics* **2006**, 100, 084501). Copyright (2006) American Institute of Physics.

The stress behaviors of these frozen-junction PLECs are very different from those of dynamic-junction PLECs operated at room temperature (RT). The EL intensity initially

experienced a sharp decline. Afterwards, both EL and driving voltage barely changed. The EL intensity, in particular, was nearly flat. A most remarkable fact about these stress tests is the extremely high current density used, up to $1,000 \text{ mA/cm}^2$ for a $50 \text{ }\mu\text{A}$ stress current. During the 165-hour stress at $50 \text{ }\mu\text{A}$, $594,000 \text{ C/cm}^2$ of charge had passed through the polymer film/junction. This corresponds to a very high degree of redox stability that is comparable to a PLED with a half-life of 20,000 hours stressed at 8.3 mA/cm^2 . [57]

For practical applications, frozen-junction PLECs need to operate at RT. Two general approaches have been explored to realize an RT frozen-junction PLEC. The first exploits electrolyte materials with a melting or glass transition above RT. The PLECs are first activated via heating or self-heating to establish doping and a light-emitting junction. The PLECs are subsequently cooled to RT to freeze the junction. RT frozen-junction PLECs have been demonstrated with crown ether-based electrolytes, [66,67] a light-emitting polyelectrolyte [68], a high T_g random copolymer-lithium complex, [69] and an ionic liquid. [40] The second approach involves chemically fixing the PLEC junction after its formation via the use of cross-linkable or polymerizable materials. [70] One method uses polymerizable ion pairs or ionic liquids to fix the junction during the charging process. [71-74] Ion-transport materials such as a liquid ion conductor containing cross-linkable methacrylate groups can also be cross-linked to immobilize the dopant ions. [41,55,70] The various chemical stabilization techniques were compared, and the best overall results were obtained when both the counterions and the ion-transport materials were polymerized using a radical-initiator compound. [75] With a photo-sensitive initiator compound, junction stabilization can also be realized “on demand” with UV exposure. [76] Finally, a stabilized p-n heterojunction has been demonstrated using a two-layer structure that consists of a conjugated cationic polyelectrolyte and a neutral, anion-trapping conjugated polymer underlayer. [77]

The various RT frozen-junction PLECs show promising device performance and junction stability. Some devices exhibit brightness and efficiency comparable to those of dynamic-junction PLECs. [41,66] Using PEO capped with methacrylate end groups and the LiTf salt as an electrolyte, highly efficient PLECs were demonstrated to last for more than 100 hours at a peak luminance of $2,200 \text{ cd/m}^2$. This excellent stress lifetime was attributed to a stabilized junction when the methacrylate groups are polymerized during operation. [42] The RT frozen junctions,

when left under an open-circuit condition, are stable for a test duration ranging from an hour [67,68], tens of hours [41,69,75,76], to two weeks.[77]

While RT frozen junctions can be realized with novel electrolyte materials, an unexpected solution also exists. Consider again the prototypical ITO/MEH-PPV:PEO:LiTf/Al cell that was cooled to create the first frozen-junction PLEC. If PEO is entirely removed from the active layer, the resulting cell is not expected to function properly. MEH-PPV, which is non-polar and has a T_g above RT [78,79], is not a solvent of LiTf salt. Surprisingly, the ITO/MEH-PPV:LiTf/Al cells can be activated with a large voltage bias to emit under either a forward or a reverse bias. The activated cells displayed strong evidence of doping and photovoltaic response that was consistent with the formation of a doping-induced junction.[80,81] More importantly, the activated state was stable at RT. As shown in Figure 4 (a), the photovoltaic polarity was reversed after the cell was activated with a reverse bias. The signs of both open-circuit voltage (V_{OC}) and a short-circuit current (I_{SC}) were consistent with the polarity of the PLEC junction formed. The resulting *positive* I_{SC} and *negative* V_{OC} did not degrade appreciatively after a total storage time of about 550 hours (Figure 4 (b)). In a green-emitting cell activated with a -21 V bias and tested subsequently with fast voltage scans between 0 and -16 V, the luminance measured at -16 V remained unchanged for 1,140 hours after an initial drop from 39 cd/m^2 to 32 cd/m^2 . These results represent the RT frozen junction with the longest shelf life.

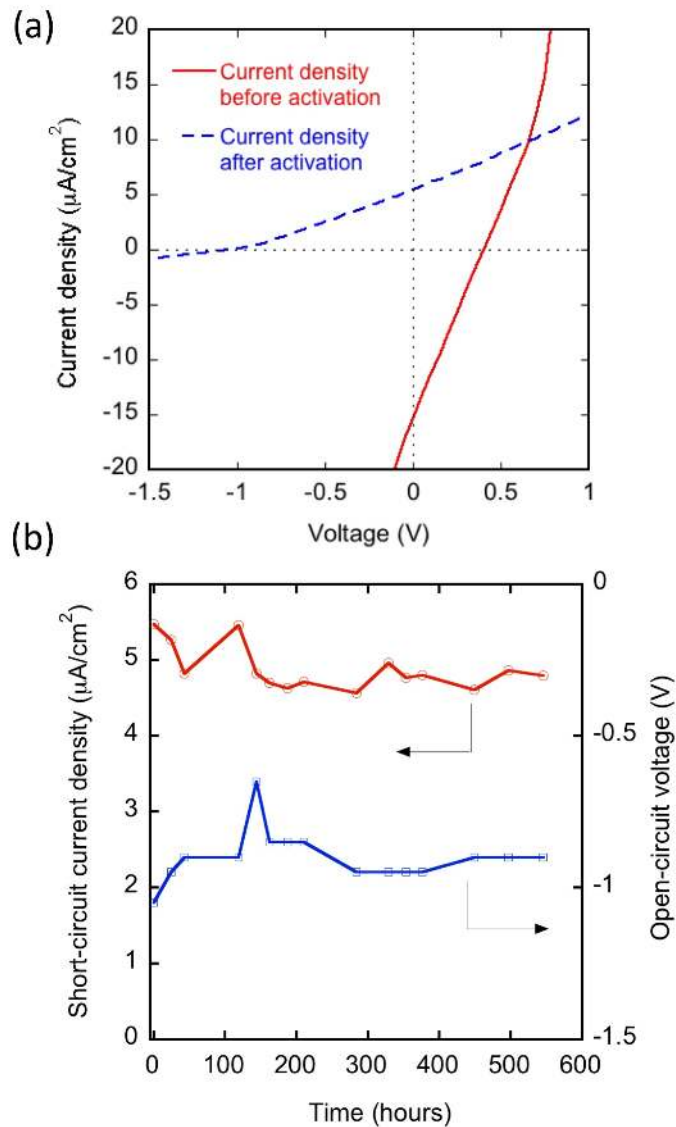


Figure 4. (a) Photovoltaic response (photocurrent vs. voltage) of a ITO/MEH-PPV:LiTf (10:1)/Al device, before and after activation under -20 V. (b) The shelf stability of V_{OC} and I_{SC} of an activated ITO/MEH-PPV:LiTf (10:1)/Al device. The device was tested under AM1.5 illumination of $100 \text{ mW}/\text{cm}^2$. For shelf stability tests, the device was stored at room temperature under open-circuit conditions. Reprinted (adapted) with permission from (Gautier, B., Wu, X., AlTal, F., Chen, S. and Gao, J., Reverse bias activation of salt-doped polymer light-emitting devices. *Organic Electronics* **2016**, 28, 47-52). Copyright (2016) Elsevier.

V. Pulsed and Intermittent Driving

The operational lifetime of PLECs can be measured with either a constant voltage stress or a constant current stress. A constant current stress is now a more commonly used method, the benefits of which are to facilitate a faster turn on and to compensate for the loss of luminance

due to any increase in cell resistance. Not surprisingly, the longest-lasting PLECs shown in Figure 1(c) were all tested with a constant current stress. Figure 5 shows generic characteristic curves of a constant current stress test. The PLEC tested was an ITO/MEH-PPV:PEO:LiTf/Al sandwich cell.[45] This continuous test yielded a luminance half-life of about 230 hours at an applied constant current density of 167 mA/cm^2 . Plotting the time axis in log scale reveals three distinct stages of the degradation process. In stage I, doping led to an increasingly balanced charge injection and an increase in luminance. The increase in luminance was accompanied by a rapid decrease in driving voltage. In stage II, the level of doping continued to increase, as evidenced by the decrease, albeit slower, of the driving voltage. The luminance, however, began its initial decline. In stage III, the decrease of luminance was accompanied by an increase in driving voltage. The first two stages are unique to LECs, while the stage III behavior is also observed in the long-term decay characteristics of a PLED.

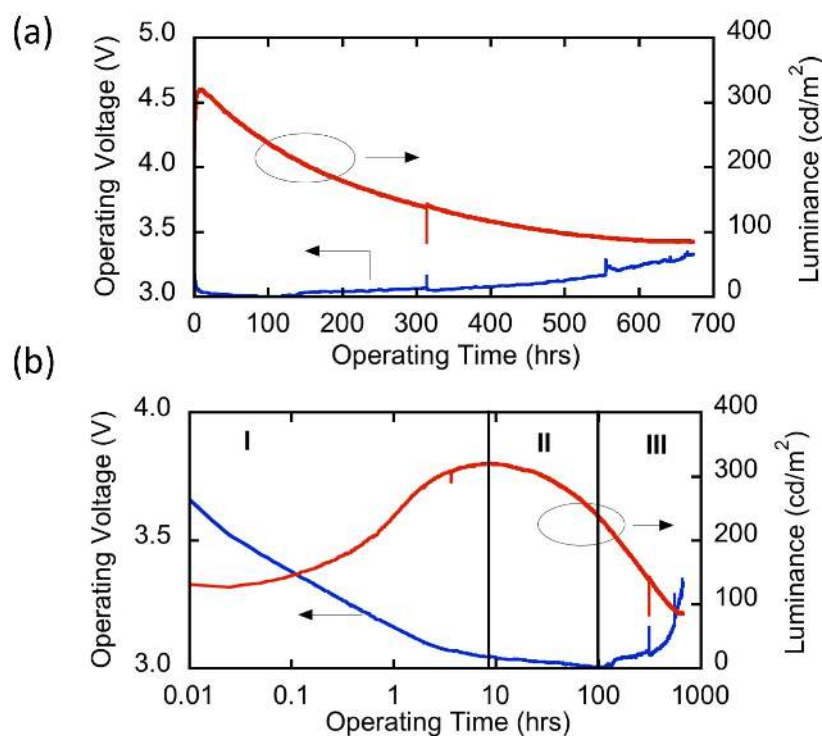


Figure 5. (a) Time evolution of luminance and operating voltage of a ITO/MEH PPV:PEO:LiTf/Al sandwich LEC (Cell 1) operated at a constant current density of 167 mA/cm^2 . The cell was operated for the first time after the deposition of the top aluminum electrode. The test commenced after the deposition of the top aluminum electrode without delay. (b) The same data presented in (a) but plotted in a semi-log form with various regions of operation marked. Reprinted (adapted) with permission from (AITal, F. and Gao, J., Long-term testing of polymer light-emitting electrochemical cells: Reversible doping and black spots. *Organic Electronics* **2015**, 18, 1-7). Copyright (2015) Elsevier.

Stage II, which lasted about 100 hours in this device and accounted for a near 20% drop in luminance from the peak value, deserves special attention. Since *in situ* electrochemical doping is known to quench the photoluminescence (PL) of PLEC films even in a sandwich cell,[82,83] is it possible that the apparent luminance decay was caused by the doping process itself? If this is the case, the loss in luminance could be reversible if the PLEC film is allowed to dedope. An intermittent testing scheme provided a definitive answer to the above question. Figure 6 shows a PLEC, similar to the one shown in Figure 5, that has been tested five times, each test lasting between 66 hours and 71 hours in each of the tests.[43] After each test (run), the cell was stored under open-circuit conduction for a month. This long storage time led to significant dedoping and the complete recovery of lost luminance in runs 2-4. In each of the five runs, the luminance decay rate increased due to the appearance of black spots and/or residue doping. The remarkable recovery of lost luminance, by simple storage, suggests that the PLEC luminance decay is not entirely irreversible!

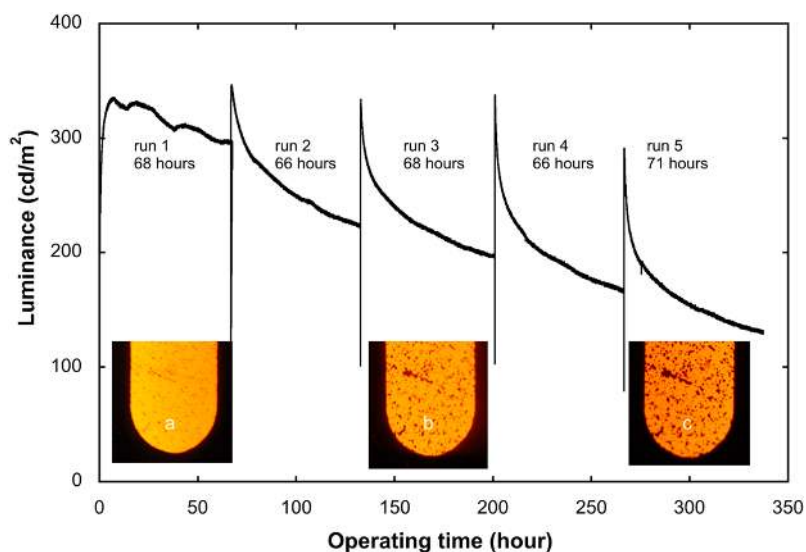


Figure 6. Time evolution of luminance and driving voltage of a ITO/MEHPPV:PEO:LiTf/Al sandwich LEC operated at a constant current of 167 mA/cm^2 . The cell was stressed five times. The operating time indicates accumulated run time under a bias. For each run, the cell was stressed for 66-71 h. The cell was stored for a month (30 ± 2 days) in the glove box at 25°C after each run. The first run (virgin run) commenced after the deposition of the top aluminum electrode without delay. Photographs a, b, and c are electroluminescent images taken at the end of runs 1, 3, and 5, respectively. Identical camera settings were applied. The images show the bottom half of the cell. They have not been manipulated except for cropping. Reprinted (adapted) with permission from (Li, X., AlTal, F., Liu, G., and Gao, J., Long-term, intermittent testing of sandwich polymer light-emitting electrochemical cells. *Applied Physics Letters* **2013**, 103, 243303). Copyright (2013) American Institute of Physics.

The intermittent testing scheme shown above resembles a pulsed driving scheme with extremely long on and off durations. A conventional pulsed driving scheme can also be applied to PLECs, where fast pulses are used to decouple the slow ionic/doping processes from the fast electronic processes. For example, a long, initial pulse can be used to activate the PLEC, and a train of short pulses can be used for the injection of electronic charges.[84] Recently, pulsed driving was applied to a 10 μm -gap planar LEC made with a green-emitting co-polymer mixed with an ionic liquid.[85] As shown in Figure 7, the 1 ms pulses had a DC offset of 3 V to activate and maintain the PLEC junction. The planar PLEC reached an extraordinarily high current density of 1,036 A/cm^2 at 30 V. By cooling the device down to $-20\text{ }^\circ\text{C}$ to freeze the junction, the PLEC showed reduced efficiency roll-off compared to RT operation. With an even narrower pulse width of 200 ns, the authors demonstrated a linear dependence between EL intensity and a record current density of 2 kA/cm^2 .

Figure 7. a) Current density–voltage characteristics of a planar LEC driven by a 1 ms voltage pulse with 3 V of base voltage. Standard DC measurements are also shown for comparison. Inset shows the same data replotted on a linear scale. b) Pulse-driving scheme for LEC. A constant base voltage, V_{base} , of 3 V was applied to induce ion arrangement for p–n homojunction formation, and a 1 ms pulse voltage, V_{pulse} , was applied to inject electrons/holes for light emission. c) Optical microscopy of emitting LECs at $V_{\text{base}} = 3\text{ V}$ and $V_{\text{pulse}} = 12$ and 30 V. d) Normalized efficiency as a function of current density at room temperature

and at $-20\text{ }^{\circ}\text{C}$, where the temperature is low enough to freeze ionic motion. The dashed line and dotted line represent $J90\%$ for polymer OLEDs in literature and for the LEC, respectively. Reprinted (adapted) with permission from (Sakanoue, T., Li, J., Tanaka, H., Ito, R., Ono, S., Kuroda, S., and Takenobu, T., High Current Injection into Dynamic p–n Homojunction in Polymer Light-Emitting Electrochemical Cells. *Advanced Materials*, 2017, 29, 1606392). Copyright (2017) WILEY-VCH.

These remarkable results exemplify the importance of a driving scheme to the operational stability of PLECs. Intermittent testing reveals that the apparent luminance decay in a PLEC is partially reversible. The combination of pulsed driving and frozen-junction operation allows for a PLEC to be driven to record current densities with minimal efficiency roll-off.

VI. Electrochemically stable emitters and electrolyte materials

Although PLEDs and PLECs often use the same luminescent CPs, the PLEC operation places additional requirements on the emitters. In PLECs, CPs must possess the requisite ability to be p- and n-doped. An early study of ten soluble PPV derivatives showed that the doping potentials, as well as the reversibility of doping were dependent on the substituent groups.[86] Another study showed that the reversibility of MEH-PPV doping depended on the electrode metals used, the salt type/concentration and film thickness.[87] These results, obtained via cyclic voltammetry measurements in a liquid electrochemical cell, are mostly consistent with the observations in PLECs.[88-92] A majority of the long-life PLECs are based on Superyellow, which can be both p- and n-doped.[51] In general, a yellow/green emitter requires a lower driving current to reach the same luminance (a photometric quantity), in comparison to a red emitter such as MEH-PPV. This means that it will be more challenging to realize long-lasting blue, red and white PLECs, although many promising materials and devices have been reported.

Unlike PLEDs, PLECs also contain an electrolyte material that is as important as the emitting material. The general requirements for PLEC electrolyte materials are: (1) the ability to contribute a sufficient amount of mobile ions required for the redox doping reaction; and (2) a wide electrochemical stability window (ESW) so as not to compete with/interfere with the doping of the CP. A study of PLECs involving six PPV-based CPs and a PEO:KCF₃SO₃ electrolyte showed that the irreversible oxidation of the electrolyte was likely responsible for the poor p-doping performance of LEPs with a high oxidation potential.[93] Moreover, cathodic side reactions involving the same electrolyte could occur in a MEH-PPV-based planar PLEC.[94] A trimethylolpropane (TMPE)-LiCF₃SO₃ electrolyte, with its superior cathodic stability, was responsible for a record operational lifetime of 1,375 hours.[52] The TMPE electrolyte was

further improved by optimizing the end group, which resulted in PLECs with a better overall performance.[6,95] Figure 8 shows the excellent performance of a PLEC made with alkyl carbonate-capped TMPE electrolytes.

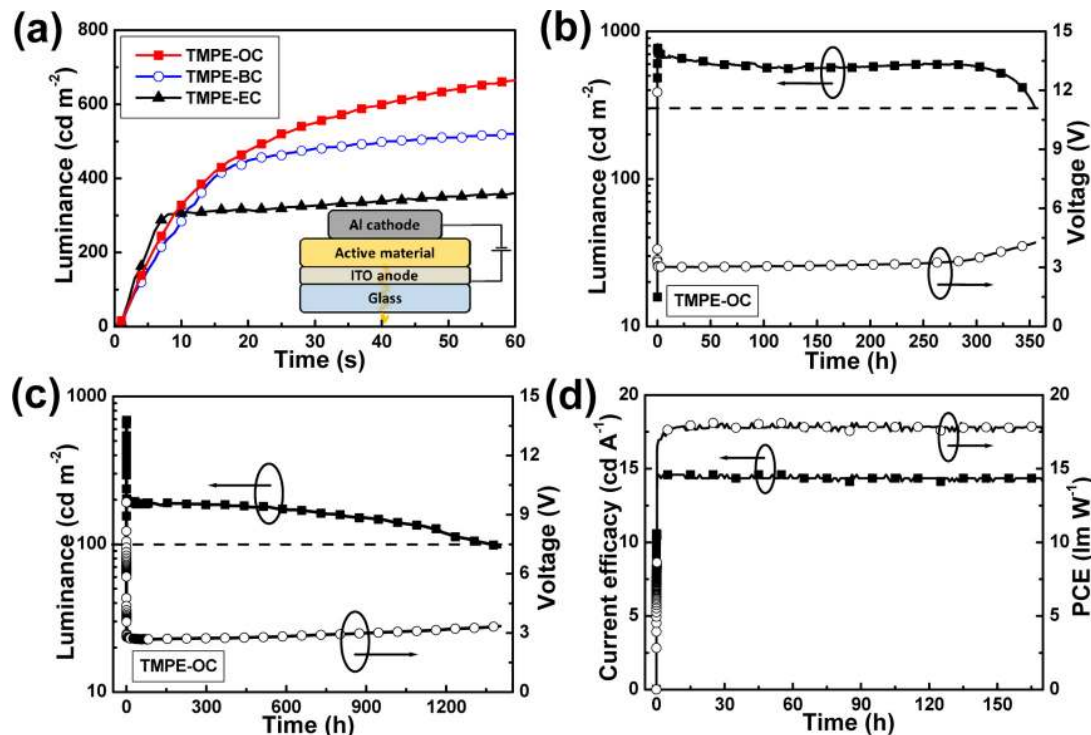


Figure 8. (a) Turn-on kinetics during the first minute of operation and (b) the long-term stability of ITO/SY + ion-transporter + LiCF₃SO₃/Al sandwich-cell LECs, with the device structure disclosed in the inset of (a). The devices were driven by $j = 7.7 \text{ mA cm}^{-2}$. (c) The long-term stability 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.; Kalthovirta, 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 miscibility of CPs and electrolyte materials is also important. In PLECs such as ITO/MEH-PPV:PEO:LiTf/Al, significant phase separation can occur between the non-polar CP and the polar PEO electrolyte. This creates a large physical separation between the CP to be doped and the counterions necessary for doping, and a consequently poor turn-on response and emission uniformity. Thermal annealing, followed by fast cooling, has been shown to effectively alleviate the problem.[96] By adding octylcyanoacetate, a surfactant-like additive to the PLEC blend, a fine, bi-continuous network morphology was realized. The resulting PLECs exhibit fast (ms) responses as well as a much-improved operational lifetime.[35] The latter work stimulated

great effort to create LECs with optimized phase morphology and the demonstration of PLECs made with crown ether (CE)-based electrolytes.[37] Being a small molecule and soluble in a non-polar solvent, CE-based electrolytes brought better compatibility with CPs and fast, efficient PLECs with a good stress lifetime. Ionic liquids (ILs), or RT molten salts, can provide the necessary counterions when added to a luminescent polymer.[97-100] The device performance of IL-based PLECs strongly depends on the type of ILs and their concentration. At high concentrations, however, the composite films again suffer from phase separation, fast degradation, and compromised mechanical integrity. An alternative is to use a polymeric ionic liquid (PIL). PLECs made with a PIL show excellent device performance with PIL loading up to 50 wt%. [101] As introduced in Section III, some ILs can also be used to realize RT frozen-junction PLECs owing to their high melting point or their ability to be polymerized.

To further improve the phase morphology of PLEC active layers, a class of bi-functional polymers has been developed. Oligo(ethylene oxide) or crown ether groups are grafted as side groups to PPVs,[102-106] polythiophenes,[107] or fluorene copolymers[108,109]. Block copolymers containing both oligo(ethylene oxide) or crown ether segments and conjugated segments were created as an alternative.[110-113] Due to the presence of ion solvating/transport moieties, these new polymers can transport both electronic and ionic charges. PLECs made with a bifunctional polymer and an added salt display characteristic LEC behaviors, such as low EL turn-on voltage and bipolar EL. Some exhibit a smoother and finer surface morphology, due to the elimination of ion-conducting polymers such as PEO.[103,106] Finally, luminescent conjugated polyelectrolytes (LCPE) [68,114-119] are multi-functional materials that are luminescent, electronically conductive, as well as being solid electrolytes. LCPEs have the advantage of being soluble in more environmentally benign polar solvents, such as water and alcohol, and allowing the fabrication of “single-component” PLECs. In general however, the device performance of these single- or two-component (polymer + salt) PLECs is inferior to conventional “three-component” PLECs. An exception is a fluorene polymer (BDOH-PF) with an ether side group.[36] When doped with LiTf salt, the resulting PLEC is highly efficient. Interestingly, adding PEO to the blend resulted in severe phase separations but a white light PLEC. Adding an iridium phosphorescent dye and lithium triflate to BDOH-PF resulted in a phosphorescent LEC which was much more efficient than PLEDs made with the same phosphorescent dye.[120]

VII. LECs with a non-polymer emitter

While the preceding discussions concern only PLECs, or LECs with a polymeric emitter, LECs have also been demonstrated with emitters as diverse as organic small molecules (SMs), [121-128] quantum dots, [129-131] polymer/small molecule composites, and even perovskites. [125,132] LECs based on ionic transition metal complexes (iTMCs), in particular, stand out due to their excellent device performance. iTMCs have been the topic of numerous recent reviews. [2,133-136] iTMCs, such as ruthenium or iridium-based complexes, are multi-functional compounds satisfying all the basic requirements of an LEC material. iTMCs are efficient phosphorescent emitters, semiconductors, as well as ion conductors. They also exhibit reversible redox properties and are solution-processible. Early reports of single-component, single-layer iTMC LECs show exceptional external quantum efficiency up to 5.5% and luminous efficiency more than 10 lm/W at low operating voltages. [137,138] Much higher efficiencies have been realized in iridium iTMC-based LECs. [139] iTMC-based LECs exhibit the same characteristic LEC transient behavior as that of PLECs. [140] The fluorescence imaging of functional planar cells confirms that iTMC-LECs, [141] along with SM-LECs [122] operate on the same basic principle as a PLEC—the electrochemical formation of a light-emitting p-n junction.

iTMCs, however, are single-ion conductors with low ionic conductivities depending on the type of counter ions, despite being single-phase systems. [137] A reflection of this fact is the absence of large, millimeter-sized planar iTMC-LECs. Planar PLECs, on the other hand, can have an interelectrode spacing of more than 10 mm or be activated with a 5 V bias voltage. [142,143] In sandwich iTMC-LECs, low ion conductivity leads to slow turn-on responses and asymmetric luminance/current vs. voltage characteristics. [138,144] The response time of iTMC-LECs can be improved with the addition of ionic species, [18,145] the use of an optimal driving scheme, the use of a low work function cathode, [138] or separate electron/hole injection layers. [146,147] The latter two approaches however, partially negate the processing and structural advantages of LECs. And in general, there is a tradeoff between the turn-on time, when improved with added ionic species, and the operating lifetime of iTMCs, although significant exceptions do exist. [145,148] In device characteristics, iTMC-based LECs bear some similarities to PLECs with a low electrolyte loading. They likewise exhibit excellent operational stability, as shown in the example below. [149]

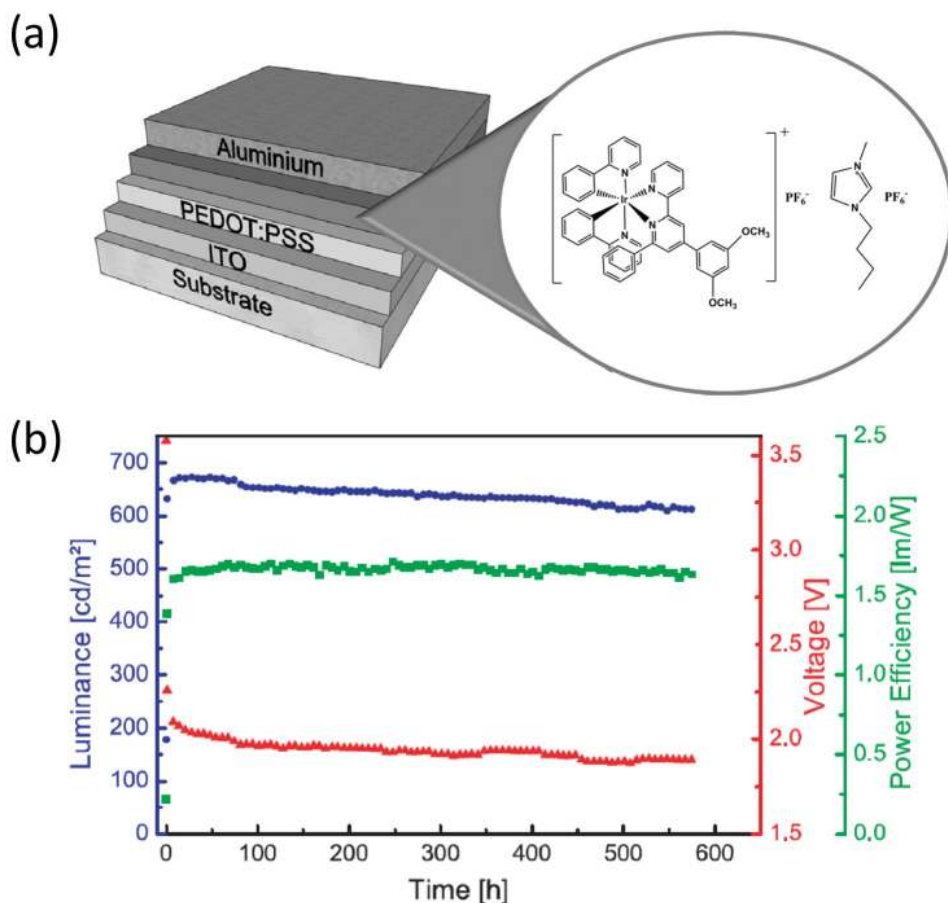


Figure 9. Layout of the LEC and chemical structures of the iTMC and the ionic liquid used in the active layer. Luminance, average voltage and power efficiency versus time for the LEC biased with a pulsed current using a block wave at a frequency of 1000 Hz and a duty cycle of 30% with an average current density of 185 A m^{-2} . Reprinted (adapted) with permission from (Tordera, D., Meier, S., Lenes, M., Costa, R., Orti, E., Sarfert, W., and Bolink, H., Simple, Fast, Bright, and Stable Light Sources. *Advanced Materials*, 2012, 24, 897-900). Copyright (2012) WILEY-VCH.

The LEC shown in Figure 9 has the structure of ITO/PEDOT:PSS/Iridium complex:ionic liquid/Al. The active layer consisted of bis(2-phenylpyridine-*C,N*)(4-(3,5-dimethoxyphenyl)-6-phenyl-2,2'-bipyridine-*N,N'*)iridium(III) hexafluorophosphate, or $[\text{Ir}(\text{ppy})_2(\text{Meppbpy})][\text{PF}_6]$ and the ionic liquid 1-butyl-3-methylimidazolium hexafluorophosphate, $[\text{BMIM}^+:\text{PF}_6^-]$ at a molar ratio of 4:1. The LEC exhibited an initial luminance of 670 cd/m^2 , which decreased to about 600 cd/m^2 during a continuous operation that lasted nearly 600 hours. A linear extrapolation yielded a T50 lifetime of more than 6,000 hours. Meanwhile, the power efficiency remained nearly constant and the (average) driving voltage decreased from over 3.5 V to less than 2 V. The excellent stability of this LEC exemplifies several effective strategies that have been commonly adopted in iTMC-LECs. First, the use of a pulsed, current driving scheme. The LEC of Figure 9

was driven with a pulsed current of 1 kHz and 30% duty cycle. The average current density was 185 A/m^2 , or 18.5 mA/cm^2 . The current drive (vs. voltage drive) enabled a fast (less than 1s) turn-on of the cell to reach significant brightness. The pulsed driving scheme was mainly responsible for improved stability vs. a DC current driving scheme, as shown in Figure 9 (d). Second, the addition of an ionic liquid additive improved the ionic conductivity and response time of the cell. Here, the particular ionic liquid did not adversely affect the stability of the LEC.[148] Third, the use of a PEDOT:PSS layer for efficient hole injection. Long lifetime iTMC-based LECs have also been reported with a modified Ru (bpy)₃²⁺ complex,[137,150], several Ir complexes, [145,151,152] BODIPY-porphyrin dyads[126].

VIII. Challenges and Opportunities

Experimental results presented in the preceding sections convey that the operational lifetime of both PLECs and iTMC-LECs have significantly improved in recent years. Long-lasting LECs are realized with three general approaches: 1. An optimized driving scheme to activate the LEC, to fix the LEC junction, and/or to drive the LEC for an extended duration; 2. An optimized LEC active layer that includes the emitting material, the electrolyte material and additives; and 3. Optimized device architecture that includes additional layers such as a hole-injection layer, a light out-coupling film, and/or the encapsulation of the LEC. Most of the longest lasting LECs, i.e., those that lasted at least 1,000 hours when operated *continuously*, [6,50,52,144,145,152-154] employ all three strategies. These strategies should also be used to optimize future LECs due to their proven effectiveness.

Increasing the operating lifetime of LECs by another one or two orders of magnitude, however, represents a major challenge. In PLECs, an increase in driving voltage, as shown in Figures 2, 5 and 8, is an often-overlooked degradation behavior. When the PLEC is driven by a constant current, an increase in driving voltage indicates an increase in the overall resistance of the cell. When the driving voltage becomes too high, rapid device destruction occurs.[49,58] In LECs, a high operating voltage can result in not only Joule heating or a dielectric breakdown, but also runaway electrochemical reactions. It is therefore, important to understand the cause of a driving voltage drift in PLECs. An unexpected observation might provide important clues. In ITO/MEH-PPV:PEO:LiTf/Al LECs with 20 % electrolyte loading such as the one shown in Figure 5, a negative (reverse bias) current stress yielded peak luminance that tripled that reached in a positive current stress.[58] The reverse bias operation, however, was less stable due to a

rapidly increasing driving voltage. When the applied reverse bias current was 83.3 mA/cm^2 , the luminance decay completely decouples from the driving voltage, as shown in Figure 10. For over 200 hours, the cell luminance stayed nearly constant between 451 cd/m^2 and 486 cd/m^2 . The driving voltage, however, increased rapidly during the same period and was likely to cause catastrophic device failure due to the level reached. These strange results suggest that the doping profile (p-dominant vs. n-dominant) plays a major role in the stability of the driving voltage. Therefore, it is important to consider both luminance decay and a driving voltage increase in any assessment of the stability of PLECs. In particular, when an extrapolated lifetime is determined, the driving voltage should also be considered. When an apparent luminance decay is caused by PL quenching, a driving voltage is a more reliable marker of true device degradation.[45]

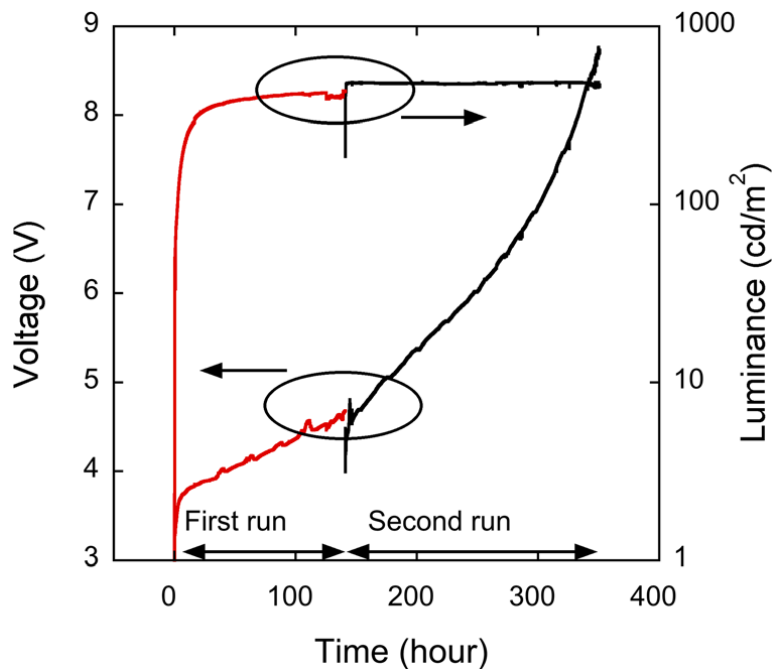


Figure 10. Time evolution of luminance and operating voltage of a ITO/MEHPPV: PEO:LiTf/Al sandwich LEC operated under a constant current of -10 mA (reverse bias). The cell is run twice with an idling period of approximately 4 days after the first run. Reprinted (adapted) with permission from (Gao, J. and AlTal, F., Decoupled luminance decay and voltage drift in polymer light-emitting electrochemical cells: Forward bias vs. reverse bias operation. *Applied Physics Letters* **2014**, 104, 143301). Copyright (2014) American Institute of Physics.

Interestingly, driving voltage increases have not been observed in some long-life iTMC LECs, as shown in Figure 8 and other studies.[145,155] When driven with a constant voltage bias, the cell current is observed to increase for over 1,000 hours.[144,152,154] This behavior has been attributed to the very low ionic mobility in these devices, causing them to experience a

prolonged activation process that obscures the decay process. Nevertheless, a stable or decreasing driving voltage represents a major advantage of iTMC-based LECs over PLECs when both are operated at RT.

The LECs with a 1,000-hour operating lifetime were all operated at low or moderate current densities. The peak luminance of these devices, however, was no less than 100 cd/m². This means that these long-lasting LECs were also very efficient. For example, a yellow-emitting PLEC operated for 1,400 hours had a power efficiency of 18.1 lm/W and a current efficiency of 14.6 Cd/A (measured at 0.77 mA/cm² on a champion device). This high efficiency allowed for the PLEC to operate at a very low current density of 1.9 mA/cm² while reaching a peak luminance of about 200 cd/m². The LEC efficiency, however, is still an order of magnitude lower than the state-of-the-art PLEDs of CDT. Although efficiency and operating lifetime are separate figures of merit concerning light-emitting devices, they appear to be closely related in LECs. Improving the efficiency, at least for PLECs, is a challenging but highly plausible route to a 10,000-hour continuous operation lifetime.

The various LEC materials can also be combined to exploit the excellent film-forming ability of polymers, the high intrinsic efficiencies of iTMCs, and the easy color tenability of small molecules and quantum dots. Indeed, hybrid LECs have been reported based on the composites of CP/QD [131], iTMC/QD,[130] SM/iTMC,[156]Polymer/iTMC[157], and PEO/iTMC[146]. Finally, novel device structures can be explored to benefit both the understanding of fundamental LEC processes and device performance. For example, LECs with coated or dispersed bipolar electrodes exhibit orders of magnitude improvement in both response times and light output due to the formation of a large-area, highly emissive “bulk homojunction.”[158,159] It would be challenging but extremely interesting to see whether the concept of bulk homojunction can be applied to sandwich LECs, although plasmonic enhancement of EL has been reported in iTMC-LECs with added gold nanoparticles.[160] At present, stacked tandem cells have been demonstrated with enhanced efficiencies or white light emission.[161-163] Significantly, when a tandem cell is operated at the same brightness as a single-layered cell made with the same emitter, it exhibits much-improved (by up to 50%) operational lifetime compared to the latter. This is due to the tandem cell’s higher external quantum efficiency, which allows it to be operated at approximately 60% of the current density of the single-layered cell to reach the same peak luminance of about 300 cd/m².[161]

IX. Conclusions and outlook

The operation lifetime of PLECs and iTMC-LECs has been surveyed. Recent advances in materials and device design have led to an operating lifetime exceeding 1,000 hours. By examining the various factors affecting the LEC operating lifetime, three general strategies have been identified as responsible for the marked improvement over early LECs. These three strategies include an optimized driving scheme, an optimized LEC active layer, and an optimized device structure. An upwardly drifting operating voltage is a major challenge in PLECs under the stress of a constant current. On the other hand, a significant portion of the luminance decay observed in PLECs is caused by a reversible doping process and is largely recoverable. Also, planar PLECs with a cryogenically fixed junction exhibit similar redox stability to a PLED made with the same luminescent polymer. To further improve the operating lifetime of LECs for practical applications, the devices must be significantly more efficient. New materials, along with innovative device structures will be the most effective way forward.

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