Self-Heating in Light-Emitting Electrochemical Cells

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Electroluminescent devices become warm during operation, and their performance can, therefore, be severely limited at high drive current density. Herein, the effects of this self-heating on the operation of a light-emitting electrochemical cell (LEC) are systematically studied. A drive current density of 50 mA cm$^{-2}$ can result in a local device temperature for a free-standing LEC that exceeds 50 °C within a short period of operation, which in turn induces premature device degradation as manifested in the rapidly decreasing luminance and increasing voltage. Furthermore, this undesired self-heating for a free-standing thin-film LEC can be suppressed by the employment of a device architecture featuring high thermal conductance and a small emission-area fill factor, since the corresponding improved heat conduction to the nonemissive regions facilitates more efficient heat transfer to the ambient surroundings. In addition, the reported differences in performance between small-area and large-area LECs as well as between flexible-plastic and rigid-glass LECs are rationalized, culminating in insights that can be useful for the rational design of LEC devices with suppressed self-heating and high performance.

1. Introduction

It is inevitable that electroluminescent devices, notably light-emitting diodes (LEDs), organic LEDs (OLEDs), and light-emitting electrochemical cells (LEC), will heat up during operation because of Joule heating,[1] the formation of dark and quenched excitons,[2] and self-absorption.[3] This self-heating is an unfortunate and problematic issue, since a too high operational temperature results in a severe drop of the device efficiency and premature device failure.[4] Accordingly, a significant number of studies on the cause and consequences of self-heating, and on how to alleviate its effects, are available in the scientific literature for both LEDs[1j–l] and OLEDs.[1a,c–i,4,5]

A LED is a compact point emitter that features a very high luminance from a small spot; it is, as such, clearly different from an OLED or a LEC, which are thin-film surface emitters. OLEDs and LECs therefore typically operate at much lower luminance levels but instead emit over a correspondingly larger area. The LEC is further distinctly distinguished from the OLED (and the LED) by its unique operational mechanism, which includes an in situ temperature-activated redistribution of mobile ions and a subsequent formation of a dynamic p–n junction doping structure.[6] It seems plausible that this LEC-specific operation could make the LEC particularly sensitive to the operational temperature,[7] and we note that Gao and co-workers report that the stability of planar LEC devices operating at low temperature is strongly dependent on the thermal conductivity of the substrate.[8]

In this context, it is surprising that the causes and extent of self-heating for practical LEC devices, and its influence on the efficiency and stability, are rarely addressed in the scientific literature.

Our goal is to remedy this shortcoming by a systematic study on the extent, effect, and alleviation of self-heating in LEC devices. We combine infrared thermography, device characterization, and simulation to establish that the active region of a common high-performance and free-standing LEC device quickly exceeds a temperature of 50 °C at a drive current density of 50 mA cm$^{-2}$, and that already this elevation in temperature induces a fast drop of the luminance and an increase of the driving voltage. We further demonstrate that this undesired self-heating can be alleviated by the design of the free-standing device so that the heat conduction to the nonemissive regions is facilitated, where the heat can be dissipated to the surroundings by convection and thermal radiation.

2. Results and Discussion

The LEC devices comprised an active material sandwiched between a transparent indium tin oxide (ITO) anode and a...
reflective Al cathode. The active material was a high-performance blend consisting of an electroluminescent conjugated polymer termed Super Yellow, a KCF,SO₃ salt, and a hydroxyl-capped trimethylolpropane ethoxylate ion-transporter. To allow for ambient-air characterization, the glass-LEC was encapsulated by two thin glass substrates, with the one on the cathode side attached with a UV-curable adhesive. The plastic-LEC was encapsulated by attaching a flexible barrier substrate on both the top and bottom side of the device using a UV-curable adhesive. For more details on the device architecture and fabrication, see the Experimental Section.

Figure 1a,b presents a schematic top-view and side-view presentation, respectively, of the glass-LEC and the plastic-LEC. The thicknesses of the top and bottom substrate and the adhesive differed somewhat for the two devices, while the thicknesses of the other device layers and the area of the substrate (L × L = 20 × 20 mm²) were identical for both devices; see Figure 1b for exact values. The emission area (l₁ × l₂) was defined by the overlap of the reflective Al electrode and the transparent ITO electrode; its size was varied in experiments that investigated the effect of the emissive fill factor (FF), which is defined as: FF = l₁ × l₂/L × L.

Figure 1c,d presents a photograph and a schematic, respectively, of the measurement setup. The devices were driven at a constant current density (j), with the compliance voltage set to 10 V, and with the ITO electrode biased as the positive anode. The LEC device was characterized in a “free-standing” horizontal position using a custom-made 3D-printed plastic holder that minimized all solid thermal connections to the surroundings. The temperature of the upper device surface (the top substrate in Figure 1b) was measured with a thermal camera. The device was positioned at a distance of 80 mm from the focal plane of the thermal camera, and at a distance of 10 mm from an optical table (see Figure 1d). A digital camera recorded photographs of the emission area during device operation at a 30° angle with respect to the device surface.

The thermal camera effectively records the temperature of the upper surface of the device, i.e., the surface of the glass substrate for the glass-LEC and the surface of the plastic substrate for the plastic-LEC (see Figure 1b). However, a vast majority of the heat generation takes place in the active material, since the voltage primarily drops over this layer (at steady state the majority of the voltage is localized over the p–n junction in the active material). It is therefore motivated to begin this study by establishing how well the measured temperature of the upper surface represents the local temperature in the active material. For this end, we have employed finite element modeling to simulate the temporal evolution of the temperature of the glass-LEC and plastic-LEC at two different drive currents and at three different locations in the device stack—at the top surface, in the active material, and at the bottom surface—and compared these results with the measured temperature at the top surface; see the Experimental Section for details on the simulation procedure.

Figure 2 presents the measured (symbols) and the simulated (lines) temperature transients of the glass-LEC and the plastic-LEC at j = 10 mA cm⁻² (Figure 2a) and j = 50 mA cm⁻² (Figure 2b). The presented temperature is the average temperature over the 8 × 8 mm² emission area (FF = 16%) in both

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measurement and simulation. For the simulation, the electrical power was input into the active material (since this is the most resistive part of the device over which a majority of the voltage drops) and it was set equal to the average measured power during the first 5 min of operation (see Figure 3a,d). We find that the simulated temperature transients for the top surface (dashed lines) replicate the measured temperature transients at the top surface (symbols) with good accuracy within $<2^\circ$C for both the glass-LECs and plastic-LECs and at both low and high drive current densities, which demonstrates that the simulation is capable of replicating the experiment.

We further call attention to the very minor ($<0.6^\circ$C) simulated temperature difference between the central active material and the two outer device surfaces (as illustrated by the separation of the line graphs in Figure 2) for both device types and current densities. This demonstrates that the temperature is essentially constant in the vertical direction, i.e., throughout the thickness of the device, for these LECs under common drive conditions. This finding is further visualized in the inset of Figure 2b, which presents the vertical temperature gradient of the plastic-LEC, including its closest ambient-air environment, at a high drive-current density of 50 mA cm$^{-2}$. We observe that the temperature

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**Figure 1.** The device structure and the measurement setup. a) A top-view schematic of the device architecture, with the lateral dimensions labeled. The central dashed square identifies the emission area, as defined by the overlap of the ITO anode and the Al cathode. b) An exploded side view of the device architecture, with the constituent layers and their thicknesses identified for the glass-LEC and the plastic-LEC. c) A photograph of the measurement setup, depicting the positions of the LEC device, the photodiode for the measurement of the luminance, the thermal camera for the measurement of the device temperature, and the digital camera for the probing of the spatial light-emission profile. d) A side-view schematic of the measurement geometry, indicating the distance from the device to the (upper) thermal camera and to the (lower) optical table. The power dissipation paths by thermal (long wavelength) and visible (short wavelength) radiation are also indicated.

**Figure 2.** The temperature transients determined by experiments (symbols) and simulations (lines). The measured temperature transients for the glass-LEC (solid blue circles) and the plastic-LEC (open orange squares) as well as the simulated transients at different locations in the device stack (top surface: dashed line; active material: solid line; bottom surface: dotted line) during constant current density driving at a) 10 mA cm$^{-2}$ and b) 50 mA cm$^{-2}$. The inset in (b) presents the simulated cross-section temperature profile of the plastic-LEC and its close ambient-air surroundings at steady state. The fill factor is 16% and the corresponding device structures are presented in Figure 1a,b.
is essentially constant throughout the thickness of the device, but that the temperature gradient increases markedly at the interface with the ambient air, i.e., at the interface with the largest thermal resistance. These findings are also supported by the analytical derivation presented in the Supporting Information.

We note that the finding of a negligible vertical thermal gradient throughout the thickness of the thin-film LECs is in agreement with previous studies on thin-film OLED devices. Qi and Forrest employed a transmission matrix model, which delivered a negligible temperature difference of 0.1 °C between the emission layer and the glass substrate of their OLED during high-brightness operation.[1h] Park et al. performed a 1D numerical simulation that established that the emergence of a significant vertical temperature gradient required a unreasonably low value for the thermal conductivity of the active material.[1f]

Importantly, we observe that the operational temperature varies markedly between the glass-LEC and the plastic-LEC for the same drive current, and we now shift our attention to the further investigation and clarification of this issue. Figure 3 presents the time evolution of the drive voltage (a,d), the average luminance (b,e) and the average device temperature (c,f) for the glass-LEC and the plastic-LEC when driven by a–c) j = 10 mA cm⁻² and d–f) j = 50 mA cm⁻².

Table 1 presents the average values for key performance metrics for a larger set of devices. All devices feature the LEC-characteristic decreasing voltage and increasing luminance during the initial constant-current operation, which confirm that they are functional LECs that exhibit in situ electrochemical doping and p–n junction formation.[1c]

We find that the minimum voltage is somewhat lower for the glass-LEC than for the plastic-LEC (2.9 vs 3.5 V at 10 mA cm⁻²; 3.7 vs 4.2 V at 50 mA cm⁻²), which we attribute to the lower sheet resistance of the ITO-on-glass electrode (20 Ω cm⁻¹) compared to the ITO-on-plastic electrode (250 Ω cm⁻¹). We further observe that the peak luminance is similar at 440–450 cd m⁻² for both devices at the lower current (Figure 3b), but that the plastic-LEC exhibits a higher peak luminance of 2500 cd m⁻² than the glass-LEC at 1800 cd m⁻² at the higher current (Figure 3e).

During the low-current driving at j = 10 mA cm⁻², the average device temperature stabilizes after a few minutes of operation at a low value of 26 °C for the glass-LEC and 31 °C for the plastic-LEC (Figure 3c), and remain at this temperature throughout the entire measurement of 8 h. We find that the drive voltage and the luminance also remain stable in this time span, with the exception of a slight decrease in the luminance between 5–30 min for the glass-LEC.

At the high-current driving at j = 50 mA cm⁻², the device behavior is radically different. Figure 3f discloses that the average temperature of the glass-LEC quickly increases to 40 °C during the first 20 min of operation, then increases slowly to 50 °C during the following 6 h, and thereafter

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Table 1. Summary of the device performance as a function of device type and drive current density. The presented values are the average within the group of investigated 2–4 devices, as detailed by the number in () in the first line.

| Device (number of devices) | Glass-LEC (4) | Plastic-LEC (2) | Glass-LEC (3) | Plastic-LEC (3) |
|----------------------------|---------------|-----------------|---------------|-----------------|
| Current density [mA cm⁻²]  | 10            | 10              | 50            | 50              |
| Minimum voltage [V]        | 2.9           | 3.5             | 3.7           | 4.2             |
| Peak luminance [cd m⁻²]    | 435           | 447             | 1830          | 2470            |
| Peak power efficacy [lm W⁻¹] | 4.4          | 4.0             | 2.9           | 3.6             |
| Peak current efficacy [cd A⁻¹] | 4.4          | 4.5             | 3.6           | 5.0             |
| Operational lifetime [h]   | NA            | NA              | 5.6           | 1.1             |

*Defined as the operational time at a luminance > 500 cd m⁻².
begins to increase rapidly again. This marked temperature increase is reflected in the device performance. After the initial (3–4 min long) doping process, the drive voltage increases steadily from 4 to 6 V during the increase of the temperature to 50 °C, after which it increases more rapidly to the voltage compliance of the measurement (10 V). The luminance decreases at a constant rate from 1800 to 670 cd m\(^{-2}\) during the same time span, and after 6 h it begins to drop at an even faster rate.

The temperature evolution of the plastic-LEC is even more dramatic, with a very fast increase to 50 °C during the first minutes of operation (when the initial doping takes place), thereafter a lowered, but still significant, increase up to 80 °C at the end of the measurement after 1.5 h of operation. The drive voltage increases with a super-linear rate and reaches the compliance of 10 V after 1.5 h, while the luminance decreases monotonically from 2500 to 555 cd m\(^{-2}\), within the same time frame.

The conclusion from these combined observations is that: i) an increase of the current density from 10 mA cm\(^{-2}\) to 50 mA cm\(^{-2}\) results in a transition from a stable close-to-ambient device operation to a temperature runaway situation; ii) the plastic-LEC in general heats up at a faster rate than the glass-LEC; and iii) an increasing operational temperature results in a simultaneous degradation of the transport properties of the active material (as reflected in the increasing voltage) and the luminance capacity of the emitter (as reflected in the lowered luminance).

We also mention that the higher peak luminance for the plastic-LEC than the glass-LEC at the higher drive current is a consistent but transient observation (see Table 1), which demonstrates that an increasing temperature can result in a short-lived improvement of the emission efficiency of the device. We speculate that this behavior is due to a temperature-induced broadening or shift of the position of the emissive p–n junction within the active material, since the LEC performance is sensitively dependent on these properties.\(^{2e,8,13}\) This junction shift/broadening could, in turn, originate in a temperature-induced change of the relative mobility of the electrons/holes (or cations/anions). We also mention that the final failure of the investigated LEC devices followed two principal scenarios: i) a gradual degradation of the active material, as manifested in a gradual increasing voltage (to the compliance limit of 10 V) and a gradual loss of luminance or ii) a sudden formation of an electrical short-circuit through the active material, as manifested in a sudden drop of the voltage and luminance to a zero value.

The presented temperature data have, so far, been spatially averaged values, and Figure 4, therefore, presents spatially resolved maps of the measured light-emission intensity (a–d) and the measured device temperature (e–h) over the entire substrate, including the emission area and the surrounding “nonemissive substrate area” (see Figure 1). The selected spatial maps

![Figure 4](https://example.com/figure4.png)

**Figure 4.** The spatial distribution of the light-emission intensity and temperature. a–d) Photographs and e–h) thermographs depicting the 2D spatial distribution of the light-emission intensity and the temperature, respectively, for a,b,e,f) the glass-LEC and c,d,g,h) the plastic-LEC at peak luminance (\(t = 1\) min) and when the luminance has dropped to half its peak value. The time evolution (indicated by the arrows) for i,j) the light-emission intensity and k) the temperature along the centerline depicted in (a) and (c) for the glass-LEC and the plastic-LEC. The profiles correspond, respectively, to \(t = 1\) min, 5 min, 30 min, 1.2 h, and 4.5 h (only for the glass-LEC). The gray region indicates the emission area. The current density was 50 mA cm\(^{-2}\), the emission area was 8 × 8 mm\(^2\), and the substrate area was 20 × 20 mm\(^2\), corresponding to FF = 16%.
were recorded at two distinct events during the high-current device operation: at peak luminance after ~1 min, and when the luminance had dropped to half its peak value. Figure 4i–k displays the time evolution of the light intensity and the device temperature along the horizontal centerline indicated by the dotted line in Figure 4a–h.

We find that the light-emission intensity is uniform and constant over the entire emission area for both the glass-LEC and the plastic-LEC at peak luminance at 1 min, but that with increasing operation time the emission intensity from the center of the device decreases whereas the edge emission remains essentially intact. This drop in the center emission intensity is notably faster for the plastic-LEC than the glass-LEC.

The spatial temperature distribution is more complex, since also the “nonemissive substrate area” that is surrounding the emission region exhibits an increase in temperature during device operation; see Figure 4k where the emission area is indicated by the gray background. In other words, heat generated in the emissive region during device operation is transferred by conduction to the nonemissive substrate area. For instance, for the glass-LEC at $t = 5$ min, the temperature at the emission center is 41 °C, the edge emission is 38 °C, and the “nonemissive substrate area” is ~30 °C. For the same drive current density and operational time, the corresponding temperature map for the plastic-LEC reveals a much higher center emission temperature of 62 °C, a slightly higher edge emission temperature between 40 and 50 °C (non-symmetric), and a cooler “non-emissive substrate area” temperature of ~27 °C.

The radically different spatial temperature profile for the glass-LEC compared to the plastic-LEC can be explained by the more efficient lateral thermal conduction in the glass-LEC. The heat conduction ($P$) in the lateral direction is described by Fourier’s law

$$P = \kappa \cdot A \cdot \nabla T = \kappa \cdot l_{1,2} \cdot I \cdot \nabla T$$

(1)

where $\kappa$ is the thermal conductivity, $A$ is the cross-sectional area, and $\text{VT}$ is the temperature gradient in the lateral direction. The glass-LEC features a ~5.5 times higher thermal conductivity than the plastic-LEC ($\kappa_{\text{glass}} = 1.1$ W m$^{-1}$ K$^{-1}$, $\kappa_{\text{plastic}} = 0.2$ W m$^{-1}$ K$^{-1}$) and a four times higher cross-sectional area (see Figure 1b), which results in that the lateral thermal conductance of the glass-LEC is ~22 times higher than for the plastic-LEC.

This implies that the heat generated in the emission zone of the glass-LEC is more efficiently transferred to the surrounding “non-emissive substrate area” so that the overall temperature profile over the entire substrate is relatively flat. For the plastic-LEC this lateral heat flux is inhibited by the comparatively low thermal conductivity and the small cross-sectional area, which result in the observed behavior of the emission area becoming hotter (particularly the center emission) and the “non-emissive substrate area” being cooler; see Figure 4k.

A lower thermal conductivity can obviously be compensated by a correspondingly larger cross-sectional area, which has been verified in simulations. We also note that our findings are in agreement with studies on OLEDs, where the temperature increase of the emission area could be suppressed by the employment of a substrate with a higher thermal conductivity. [3c,5a] We also emphasize that our measurements were performed on “free-standing” LEC devices (see Figure 1d), and that a solid structure with a significant heat capacity and surface area (i.e., a conventional heat sink) in thermal contact with the bottom part of the device will perform a similar heat-removal function and in the process keep the device temperature at a lower value.

We now turn our attention to the connection between temperature and light intensity. Importantly, the recorded data consistently imply a direct correlation between an increase in the local temperature and a drop in the local light intensity, which suggests that the degradation of the emission capacity is thermally activated. More specifically, Figure 4i reveals that the light intensity is uniform over the entire emission area for the glass-LEC at $t = 1.2$ h, but that at $t = 4.5$ h, the center emission has dropped significantly, while the edge emission is practically retained. The correlation with the temperature traces displayed in Figure 4k suggests that a stable long-term operation is prohibited already at a modest operational temperature of ~45 °C.

For the plastic-LEC, the light emission is only uniform during the first 5 min of operation, and thereafter the center emission starts to drop, while the edge emission again remains constant up to the end of the measurement at $t = 1.2$ h; see Figure 4j. The correlation with the temperature traces in Figure 4k indicates that the “threshold” for stable long-term operation of the plastic-LEC is positioned at a slightly higher temperature of ~50–55 °C. However, it should be noted that a minor degradation of the luminance capacity of the plastic-LEC at a lower temperature most likely is obscured by the quick temperature increase in this measurement. Nevertheless, it is clear that a prolonged operation above 50 °C has a negative impact on the stability of the emission capacity for both the glass- and plastic-LECs (although a short high-temperature stint can actually boost the efficiency, as shown in Figure 3e,f).

We also comment on the evolution of the shape of the temperature profiles in Figure 4k. The initial Gaussian shaped temperature profile is much flattened at the end of the measurement, which we attribute to a spatially selective degradation of the hotter center region. The degraded central part becomes more resistive, which in turn lowers the current density through the (more resistive) central region and increases the current density through the (less resistive) edge regions in order to maintain an overall constant current density.

All investigated LEC devices, up to this point, featured an invariant emissive fill factor (FF) of 16%, as calculated by dividing the $8 \times 8$ mm$^2$ emission area by the $20 \times 20$ mm$^2$ substrate area. In order to investigate the influence of the FF on the device performance and the device temperature, we have in addition fabricated and evaluated glass-LECs with emission areas of $2 \times 8$ mm$^2$ and $12 \times 16$ mm$^2$ (at a retained substrate area of $20 \times 20$ mm$^2$), which accordingly featured an FF of 4% and 48%, respectively.

Figure 5a–c presents the temporal evolution of the device performance of the three glass-LECs with FF = 4% (glass-LEC4%, solid green triangles), 16% (glass-LEC16%, solid blue circles), 48% (glass-LEC48%, red diamonds), and a plastic-LEC with FF = 16 (plastic-LEC16%, open orange squares) during operation at $j = 50$ mA cm$^{-2}$. Figure 5a shows that the minimum voltage is slightly higher for glass-LEC48% and plastic-LEC16%, which is attributed to the larger ITO area and its
higher resistance. Figure 5b displays that the peak luminance is higher at ≈2200 cd m\(^{-2}\) for the glass-LEC48\% and plastic-LEC16\% in comparison to ≈1900 cd m\(^{-2}\) for the glass-LECs with smaller FF. A comparison with the temperature transients in Figure 5c reveals that this short-lived high luminance is delivered at a high average device temperature of >50 °C, which yields support for our previous claim that an increase in temperature can (but not necessarily) shift the emissive p–n junction to a more efficient position in the active material\[^{2e,13a–c}\] or alternatively broaden it for lowered exciton quenching\[^{8,13e,f}\].

We further find that the average device temperature is strongly dependent on the FF, with the largest-FF glass-LEC48\% surpassing 60 °C already after 2 h, the intermediate-FF glass-LEC16\% reaching 50 °C at 6 h, while the smallest-FF glass-LEC4\% remains below 35 °C throughout the entire measurement period of 8 h. The device data in Figure 5a–c thus yield further support for our finding of a strong negative correlation between the device temperature and the operational stability of LEC devices, as exemplified by that the luminance of the hottest glass-LEC48\% drops below 1000 cd m\(^{-2}\) already after 2 h, while the lowest-temperature glass-LEC4\% emits above 1000 cd m\(^{-2}\) for the entire measurement period of 8 h. Importantly, this finding has the implication that a report on LEC stability always should be accompanied by a detailed description of the device structure and the measurement geometry.

Figure 5d presents the measured increase in temperature of the LEC device averaged over the emission area (\(\Delta T\)) as a function of the power density, i.e., the input electrical power (\(P\)) divided by the emission area (\(A_{\text{emission}}\)). We emphasize that \(\Delta T\) is essentially identical to the difference between the average temperature of the active material (which is essentially the same as the average temperature of the LEC device) and the measured initial temperature when the device was in thermal equilibrium with the ambient (≈22 °C). Figure 5d shows that the measured data for the four different LEC devices (as identified in the inset) fall on top of four different straight lines, which suggests that the following linear equation for the heat transfer between a LEC device and the ambient air holds:

\[
\Delta T = \left( \frac{1}{h} \right) \frac{P}{A_{\text{emission}}} \tag{2}
\]

where \(h\) represents an effective heat transfer coefficient, which includes all the dissipation mechanisms—i.e., a combination of convection, radiation, and conduction. Accordingly, the value for \(h\) for a particular LEC device can be derived as the inverse
of the slope of its graph in Figure 5d. We find that the value of $h$ is 63.7 $W \cdot m^{-2} \cdot K^{-1}$ for the glass-LEC$_{48\%}$, 130 $W \cdot m^{-2} \cdot K^{-1}$ for the glass-LEC$_{16\%}$, 339 $W \cdot m^{-2} \cdot K^{-1}$ for the glass-LEC$_{4\%}$, and 64.3 $W \cdot m^{-2} \cdot K^{-1}$ for the plastic-LEC$_{16\%}$.

The observed decrease in $h$ with increasing FF (i.e., the emission area for this study with a constant substrate area) is due to a combination of two effects: i) the input electrical power (specifically the current) increases linearly with increasing emission area at constant current density. The same is true for the heat dissipation from the device area by direct convection and radiation, but not for the lateral heat conduction, which instead is linearly proportional to the cross-sectional area of the device. Since the device thickness is constant, the lateral heat conduction scales only with the emission-area perimeter. The consequence is that an increasing FF results in an increase of the input electrical power (and the direct heat dissipation by radiation and convection) by a factor $\propto l_1 \times l_2$ (see Figure 1b for definition of $l_1$ and $l_2$), while the lateral thermal conduction increases with a slower rate $\propto (l_1 + l_2)$. ii) With increasing FF, the “non-emissive substrate area” decreases, which means that the subsequent dissipation of the heat transferred to this area by convection and radiation decreases. These two effects will both result in a lower value for $h$ with increasing FF, as manifested in a higher device temperature. Figure 5e,f provides a schematic illustration of the decrease of the lateral conduction to the “non-emissive substrate area,” and the correlated drop in convection and thermal radiation from this area, with increasing FF.

The lower value for $h$ for the plastic-LEC$_{16\%}$ device compared to the glass-LEC$_{16\%}$ device is due to the lower thermal conductance of the former, which inhibits the lateral heat conduction, as discussed previously and as schematically depicted by a comparison of Figure 5e,g. In fact, we note that the value for $h$ is essentially identical for the glass-LEC$_{48\%}$ and plastic-LEC$_{16\%}$ devices, which demonstrates that the 22 times lower thermal conductance at an application-relevant larger FF should, however, be considered in the light of that a larger-area OLED or LEC devices obviously can be run at a lower light intensity (and a corresponding lower power density) in order to produce the same total light-flux output. For large-area lighting applications, it is also plausible that an appropriate heat sink can be positioned in good thermal contact with the substrate barrier below the reflective electrode; such a heat sink will efficiently increase the value of $h$, so that large-FF LEC and OLED devices can be run at a reasonable operating temperature at large electrical power input. It should finally be mentioned that free-standing patterned-emission applications of LECs and OLEDs obviously can be effectuated at low values for FF.\[14\]

3. Conclusions

We demonstrate that LEC devices can self-heat to high temperatures during light-emission, and that a common high-performance LEC degrades rather rapidly when the local operation temperature exceeds 50 °C. We further report that this undesired self-heating is highly sensitive to the device architecture and that it can be strongly suppressed by rational design. Specifically, we show that an increase of the thermal conductance and a decrease of the emission-area fill factor result in much improved thermal dissipation and significantly enhanced stability for a free-standing LEC. Our findings further rationalize observed differences in performance between small-area and large-area LEC devices and between flexible-plastic and rigid-glass devices. The study also highlights the necessity of being stringent and consistent when reporting the conditions of device characterization, and finally provides guidelines for how LEC devices are to be designed for optimal operation in specific applications.

4. Experimental Section

The LEC devices were fabricated as follows. The indium tin oxide (ITO)-coated glass substrate (Thin Film Devices Inc, US) and the ITO-coated plastic substrate (poly(ethylene terephthalate), PET) were cleaned by sequential ultrasonic treatment in detergent (Extran MA 01, Merck), DI-water, acetone, and isopropanol under ambient conditions. The sheet resistance was 20 $\Omega \cdot \square^{-1}$ for ITO on glass and 250 (±100) $\Omega \cdot \square^{-1}$ for ITO on plastic, as measured by a four-probe setup. The active material comprised a blend of an electroluminescent conjugated phenyl-substituted poly(paraphenylene vinylene) copolymer termed Super Yellow (SY, Merck, Darmstadt, DE), a KCF$_3$SO$_3$ salt (Alrich, and a hydroxyl-capped trimethylpropane ethoxylate (TMPE-OH, Alrich; $M_w = 450$ mol$^{-1}$) ion-transporter. Master solutions were prepared with cyclohexane (Alrich) as the solvent, and the following solute concentrations: 8 g L$^{-1}$ (SY), 10 g L$^{-1}$ (KCF$_3$SO$_3$, and TMPE-OH). The blend ink was prepared by mixing the master solutions in a solute mass ratio of SY:TMPE-OH:KCF$_3$SO$_3 = 1:0.15:0.03$.

The blend ink was spin-coated on the ITO-coated substrate at 2000 rpm for 60 s. The spin-coated film was dried at 70 °C for 2 h, and the thickness of the dry active material was 150 nm, as measured by a stylus profilometer (Dektak). The reflective Al electrode was deposited on top of the active material by thermal evaporation under vacuum ($p < 8 \times 10^{-4}$ mbar), and its area was defined by a shadow mask. To allow for ambient-air characterization, the glass-LEC was encapsulated by attaching a thin glass substrate on top of the Al electrode with a single-component and UV-curable epoxy (Ossila).\[10\] The plastic-LEC was encapsulated by attaching a flexible barrier substrate (Fraunhofer FEP) on both the top and bottom sides of the device using a UV-curable adhesive (Lumtec). Accordingly, the top “substrate” for the plastic-LEC in Figure 1b comprises the PET film, the adhesive, and the flexible barrier. The water vapor transmission rate (WVTR) of the flexible barrier was $5 \times 10^{-4}$ g m$^{-2}$ per day at 38 °C and 90% relative humidity, as measured by tunable diode laser absorption spectroscopy (HiBarSens). The oxygen transmission rate (OTR) was below the detection limit of the measurement equipment. The low values for the WVTR and OTR...
ensured that ambient moisture and oxygen were not influencing the measurement results. More details on the device fabrication are available in ref. [9a].

Figure 1a,b presents a schematic top-view and side-view presentation, respectively, of the glass-LEC and the plastic-LEC. The thicknesses of the top and bottom substrate and the adhesive differed somewhat for the two devices, while the thicknesses of the other device layers and the area of the substrate \((L \times L = 20 \times 20 \text{ mm}^2)\) were identical for both devices; see Figure 1b for exact values. The emission area \((l_1 \times l_2)\) was defined by the overlap of the reflective Al electrode and the transparent ITO electrode; its size was varied in experiments that investigated the effect of the fill factor (FF), which was defined as: \(FF = l_1 \times l_2 / L \times L\).

Figure 1c,d presents a photograph and a schematic, respectively, of the measurement setup. The devices were driven by a constant current density \((j)\) of either 10 or 50 mA cm\(^{-2}\), with the compliance voltage set to 10 V, and with the ITO electrode biased as the positive anode. A sourcemeter (Keithley 2400) was used to supply the current and record the electrical performance; and the luminance was measured within a collection cone of 5.5° and at an angle of 30°, using a photodiode equipped with an eye-response filter (BPW 21, Osram Semiconductors), which had been calibrated with a luminance meter (Konica Minolta LS-110). The temperature of the upper device surface (the top substrate in Figure 1b) was measured with a thermal camera (FLIR A300, 4x close-up lens, accuracy \(\pm 2.5 \, ^\circ \text{C}\)).

All measurements were performed on pristine devices, and 2–4 area during device operation at a 30° angle of view. When no spatial information was provided, the operational temperature of the LEC device was the average temperature over the entire emission area. A USB-controlled camera (Dine-Lite Digital Microscope) recorded photographs of the emission area during device operation at a 30° angle with respect to the device surface. All measurements were performed on pristine devices, and 2–4 independent devices were characterized for each device type and for each value of \(j\) and FF to confirm repeatability.

The finite element simulations were performed with the Comsol Multiphysics software (version 5.4), using the “Heat transfer in Solids and Fluids” and “Laminar Flow” modules connected with the “Nonisothermal Flow” module. The multilayer LEC device was modeled with the dimensions and material properties specified in Figure 1a,b, and “Nonisothermal Flow” module. The temperature of each location in the simulation, and the velocity of the air to/from the surrounding box. The internal interfaces within the LEC device included convection and conduction to the surrounding air and thermal radiation to/from the surrounding box. The boundary conditions at the external interfaces of the LEC device included convection and conduction to the ambient temperature of the walls of the enclosure box were set to 22 °C. During device operation, the electric power was input into the emission layer of the LEC. At \(t = 0\) s, the heat generation in the emission layer was initiated and the temperature of each location in the simulation, and the velocity of the air driven by buoyant convection, was calculated and tracked over time until the surface of the LEC device reached an (approximately) constant temperature (after 500 s of operation for the glass-LEC and 300 s for the plastic-LEC).

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

heat management, light-emitting electrochemical cells, self-heating, substrate properties, temperature-dependent performance

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