

Supplementary Information for:

Understanding the Superior Temperature Stability of Iridium Light-Emitting Electrochemical Cells

Melanie H. Bowler,^a Tianle Guo,^a Lyndon D. Bastatas,^a Matthew D. Moore^b, Anton V. Malko^a and Jason D. Slinker^a

^a Department of Physics, The University of Texas at Dallas, 800 W. Campbell Rd, Richardson, TX 75080, USA.

^bDepartment of Chemistry, The University of Texas at Austin, Austin, TX 78712, USA.

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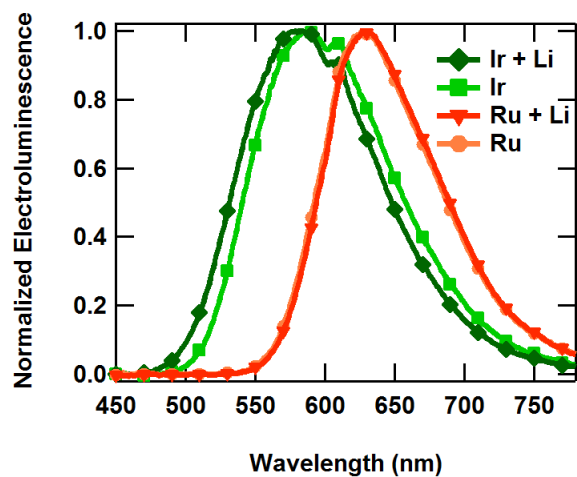
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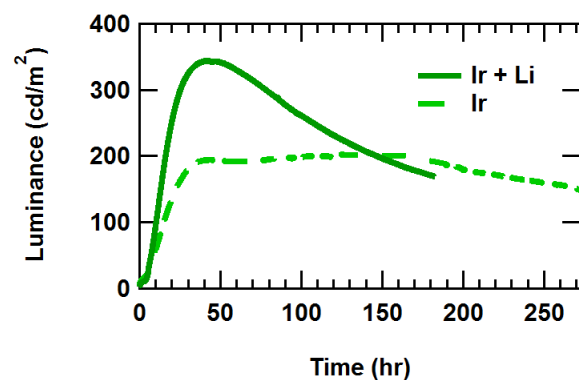
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Electroluminescence Spectra of the Devices



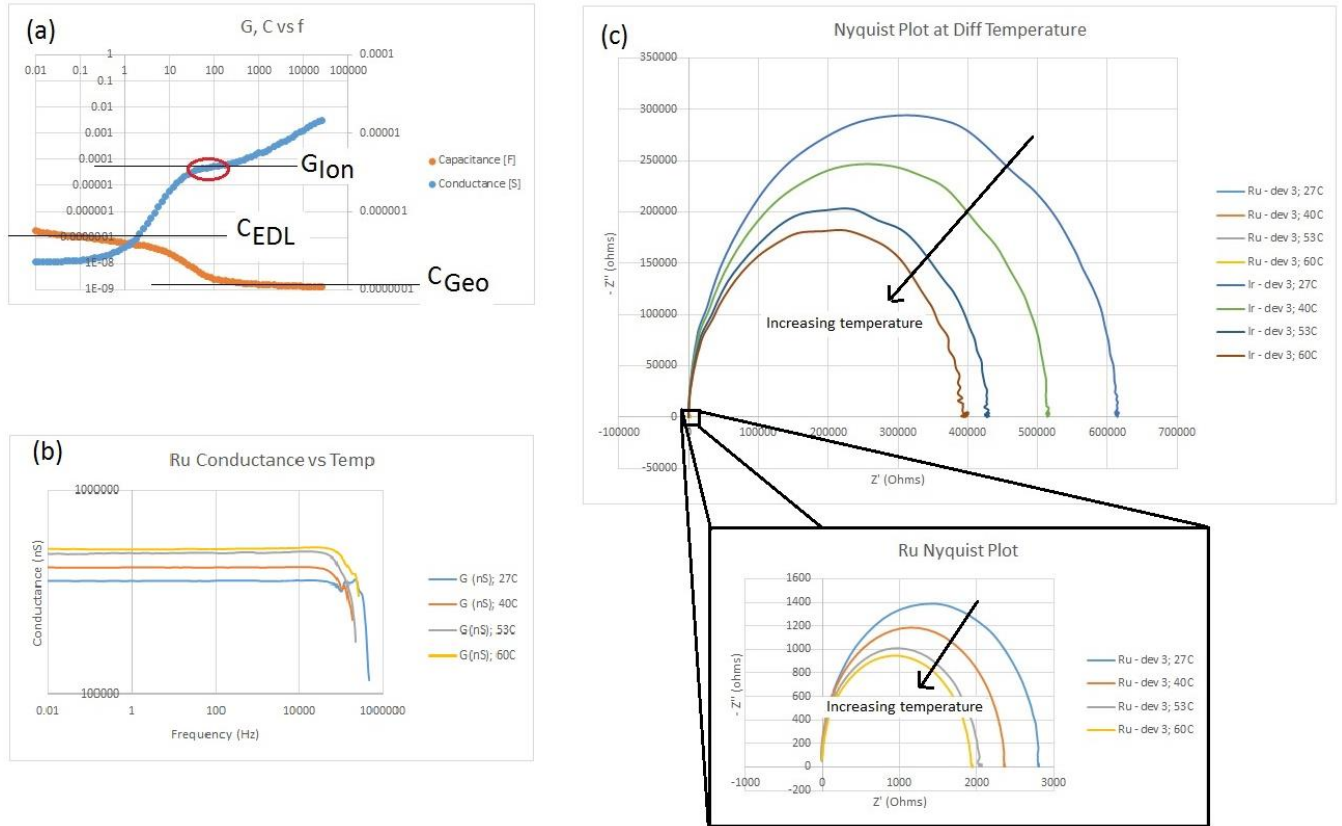
Supplementary Information Figure S1: Electroluminescence spectra for single layer LEEC devices based on $[\text{Ir}(\text{ppy})_2(\text{bpy})][\text{PF}_6]$ and $[\text{Ru}(\text{bpy})_3][\text{PF}_6]_2$, with and without 0.50%/wt $\text{Li}[\text{PF}_6]$. Salt addition does not strongly affect emission color.

Device Operation under Pulsed Driving



Supplementary Information Figure S2: Electroluminescence spectra for single layer LEEC devices based on $[\text{Ir}(\text{ppy})_2(\text{bpy})][\text{PF}_6]$ with and without 0.50%/wt $\text{Li}[\text{PF}_6]$ under pulsed current driving at 1.5 mA, 0.25 kHz and 30% duty cycle. Peak power efficiency is approximately 1 Lm/W for the iridium devices with salt.

Determining Film Conductivity through Impedance Spectroscopy



Supplementary Information Figure S3: (a) Conductance and capacitance versus frequency for a representative iridium LEEC. (b) Conductance versus frequency for a representative ruthenium LEEC. (c) Niquist plots for single layer LEEC devices based on $[\text{Ir}(\text{ppy})_2(\text{bpy})][\text{PF}_6]$ and $[\text{Ru}(\text{bpy})_3][\text{PF}_6]_2$, with and without 0.50%/wt $\text{Li}[\text{PF}_6]$.

The impedance, Z , can be decomposed into real, Z' , and imaginary part, Z'' . Symbolically, it is given as

$$\frac{1}{Z(\omega)} = \frac{1}{Z'(\omega) + iZ''(\omega)} = G(\omega) + i\omega C(\omega) \quad \text{Eq. S1}$$

where ω is the angular frequency, G is the conductive and C the capacitive response of the system under study. From Eqn. (1), one can read out the conductance as

$$G(\omega) = \frac{Z'(\omega)}{Z'^2(\omega) + Z''^2(\omega)} \quad \text{Eq. S2}$$

Figure S2 shows representative IS spectra. Following the reasoning of S. van Reenen, *et al.*, and S. Meier, *et al.*, the ion conductivity is determined when the conductive part shows intermediary plateau in regime where there is transitional increase in capacitive component that signifies electric double formation (15 Hz – 150 Hz in Figure S2a). However in some cases in this study, especially for more conductive films, such intermediary plateau in conductance does not exist but rather exhibits almost constant value that extends in wide range of frequency (Figure S2b). The G_{ion} is taken as the average of these non-deviating values. Once G_{ion} is identified, the average ionic conductivity is finally calculated as

$$\sigma_{\text{ion}} = \frac{G_{\text{ion}} d}{A} \quad \text{Eq. S3}$$

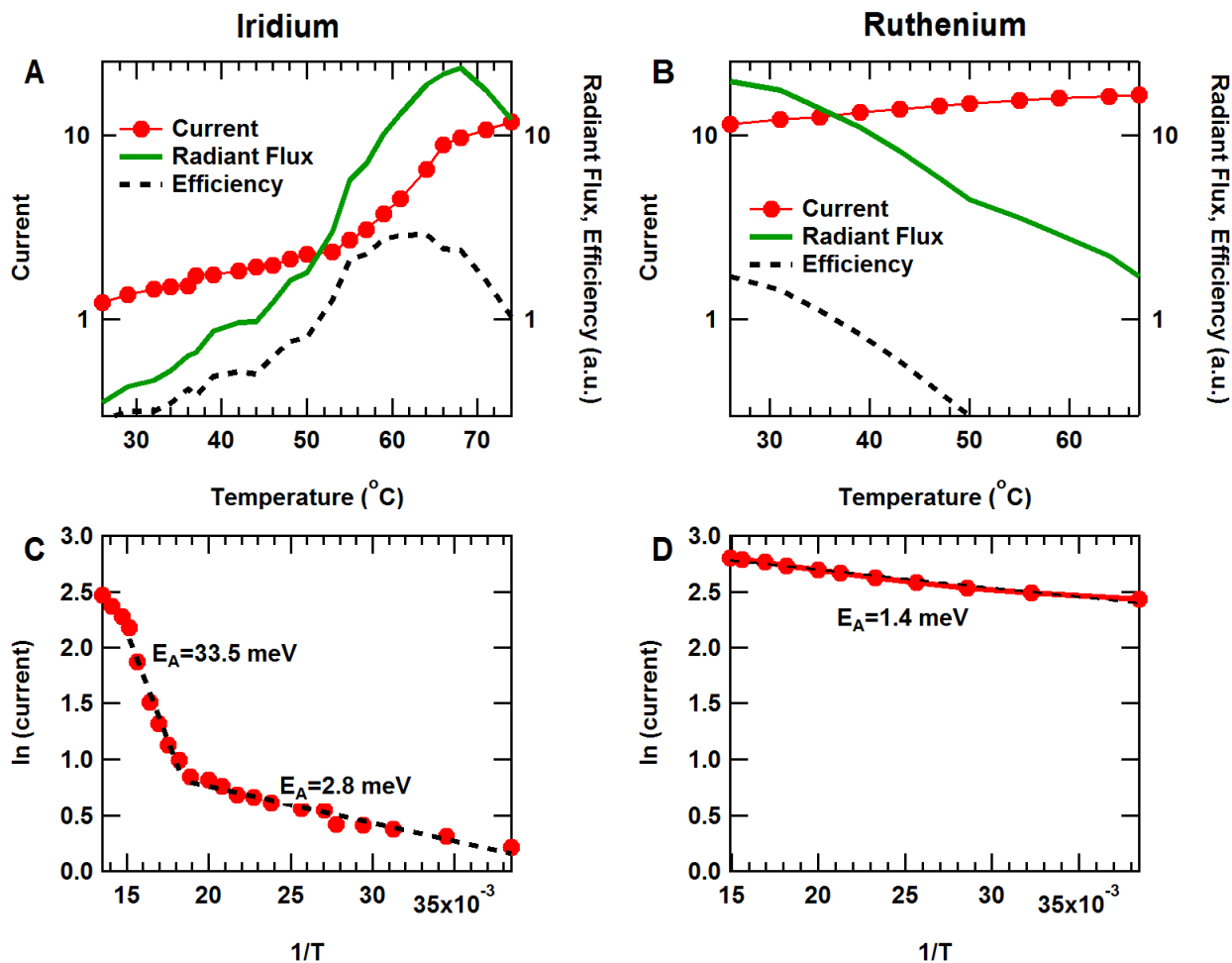
where d is the film thickness and A is the area of active region. Figure S2c is the Nyquist plot of the devices at different temperature. As can be seen, increasing the temperature in the range considered makes the film more conductive. Likewise, the bulk resistance in ruthenium film is lower than iridium.

References:

Meier, S. B.; Hartmann, D.; Winnacker, A.; Sarfert, W. The Dynamic Behavior of Thin-Film Ionic Transition Metal Complex-Based Light-Emitting Electrochemical Cells. *J. Appl. Phys.* 2014, 116, 104504.

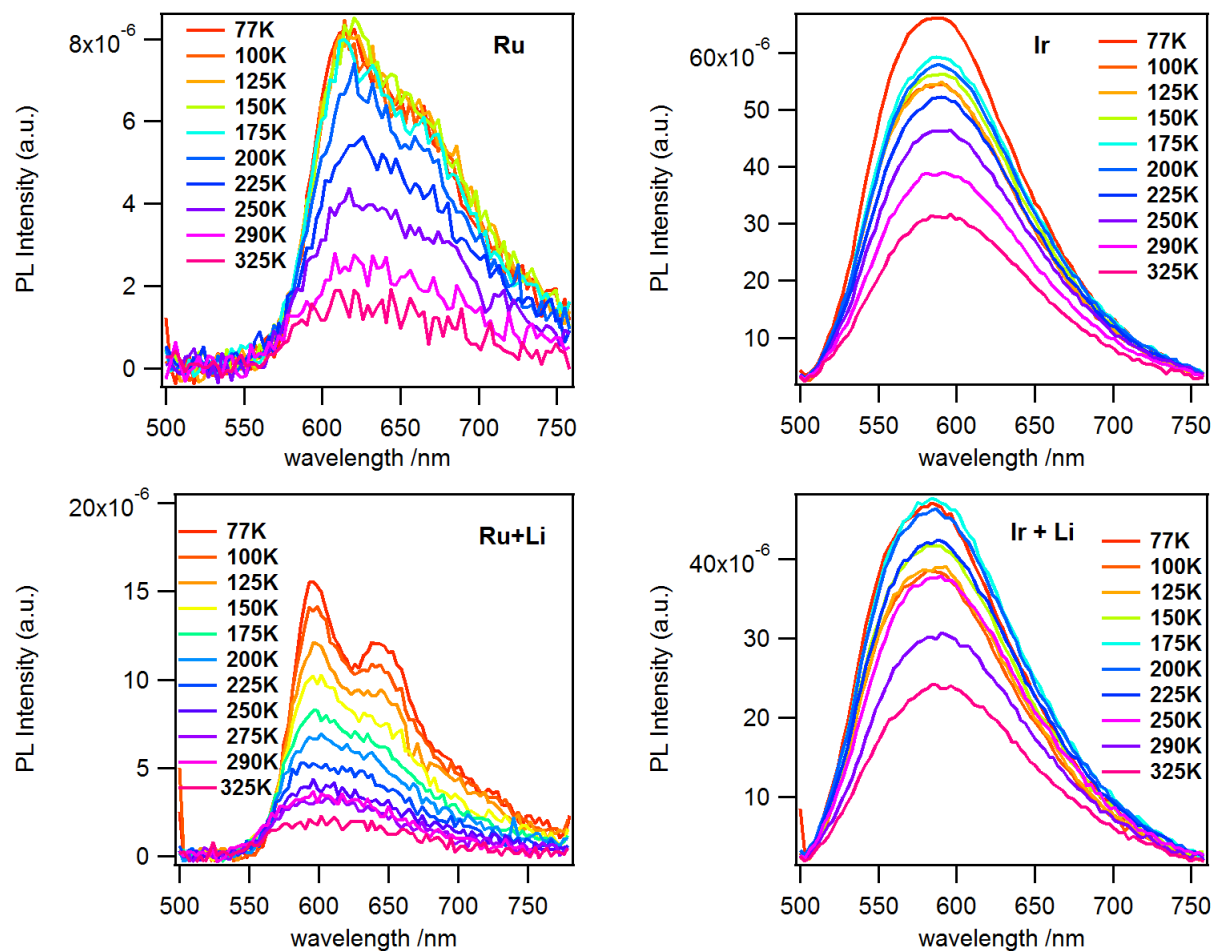
Stephan van Reenen , René A. J. Janssen , and Martijn Kemerink , Dynamic Processes in Sandwich Polymer Light-Emitting Electrochemical Cells, *Adv. Funct. Mater.* 22, 4547 (2012).

Device Temperature Dependence under Constant Voltage Driving



Supplementary Information Figure S4: Current, relative radiant flux, and relative efficiency (radiant flux/current) versus temperature of $[\text{Ir}(\text{ppy})_2(\text{bpy})][\text{PF}_6]$ and $[\text{Ru}(\text{bpy})_3][\text{PF}_6]_2$ devices with 0.50%/wt $\text{Li}[\text{PF}_6]$ under constant current (5 V and 3.5 V, respectively).

Photoluminescence Spectra for Various Temperatures



Supplementary Information Figure S5: Photoluminescence spectra for films of $[\text{Ir}(\text{ppy})_2(\text{bpy})][\text{PF}_6]$ and $[\text{Ru}(\text{bpy})_3][\text{PF}_6]_2$, with and without 0.50%/wt $\text{Li}[\text{PF}_6]$. Salt addition does not strongly affect emission color.