

## Supplementary Information

### Insights into the Degradation Mechanism of Carbene-Metal-Amide Organic Light-Emitting Diodes

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## Experimental

### Evaporation

Films were deposited using an Angstrom Engineering EVOVAC 700 system. Deposition pressures were below  $2.5 \times 10^{-6}$  mbar, but could be as low as  $5 \times 10^{-8}$  mbar. Organic materials were heated in either aluminium or alumina crucibles, while inorganic electrode materials were heated in open tungsten or graphite boats. Substrates were positioned 50 cm from the evaporation sources and rotated at 25 rpm to ensure uniform deposition. Deposition rates were generally between 0.1 and 2.0  $\text{\AA s}^{-1}$  and monitored with quartz crystal microbalances. The microbalances were calibrated for each material by measuring the thickness of test films with ellipsometry. Materials can be simultaneously evaporated, with deposition rate of each source independently monitored, allowing for the fabrication of blended films.

### Device Characterisation

Devices were electrically driven by a Keithley 2400 source meter. Current density-voltage sweeps were measured from low to high voltage. The electroluminescence of OLED devices were measured on axis using a calibrated Ocean Optics Flame spectrometer. The brightness of devices was measured with an amplified 1 cm<sup>2</sup> silicon photodiode. The output current from the photodiode was measured using a Keithley 2000 digital multimeter. Measurements were made on axis at a distance of 15 cm from the face of the device.

### Photodegradation

Degradation was studied by observing the change in photoluminescence. Samples were excited with a 405 nm laser. The laser was temperature controlled and run in constant power mode. The profile of the laser beam was constrained with an iris as close to the sample as was practical. An adjustable neutral density filter was placed between the iris and the sample, allowing for adjustment of the beam power incident onto the sample. The beam was partially reflected off the neutral density filter. The power of the reflection was monitored during the measurement with a Thorlabs S130C power meter. As the reflected power is proportional to the transmitted power, any fluctuations in excitation power can be monitored from the reflected power. Adjustments to the measured photoluminescence intensity can then be made to account for power fluctuations.

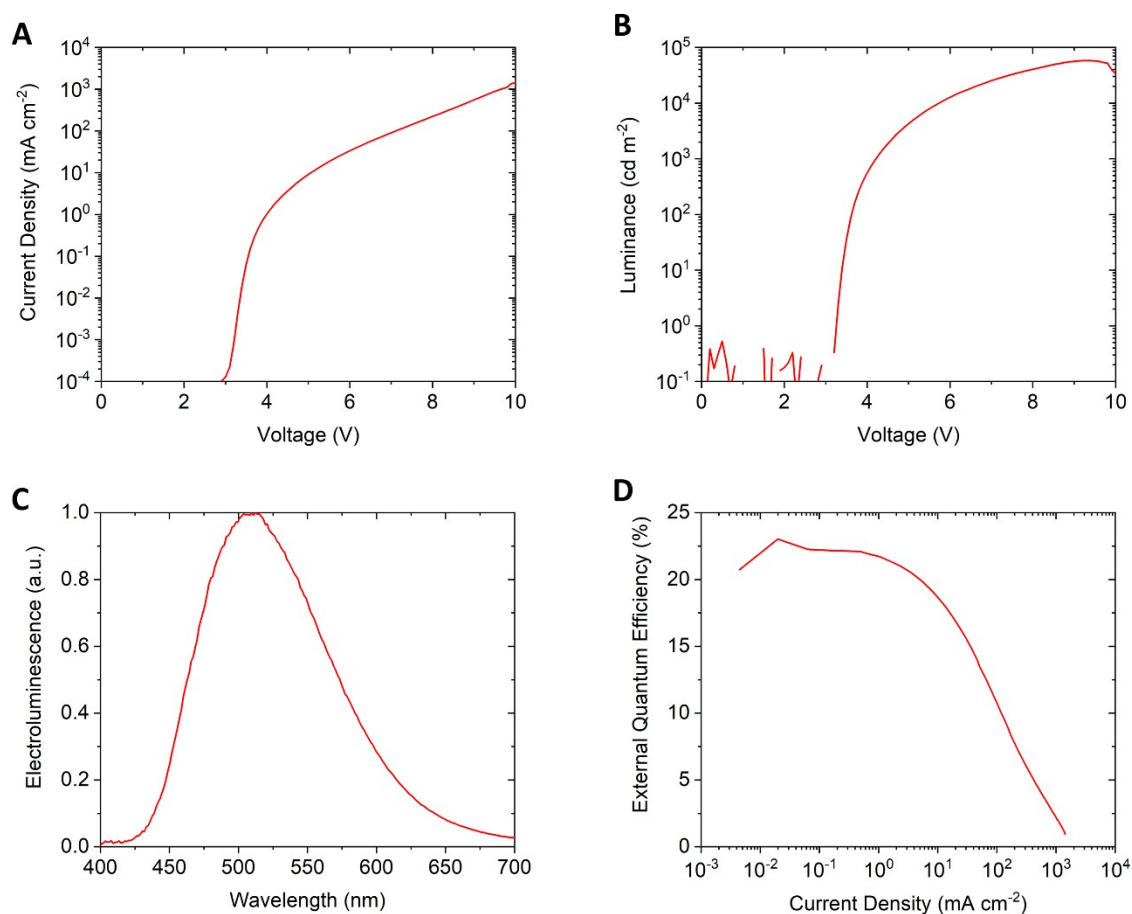
Before measurements a Thorlabs BC106N-VIS/M camera beam profiler was used to measure the beam spot at the sample position. The size, shape and power of the beam can be adjusted with the iris and adjustable neutral density filter. The iris is particularly important as it is used to cut off the outer edge of the beam where intensity drops off. A relatively uniform power density can therefore be achieved. In general the spot was about 1 mm in diameter.

The photoluminescence from the sample was collected by a pair of lenses and focussed onto an optical fibre, coupled to an Andor Shamrock spectrometer and Andor iDus CCD array.

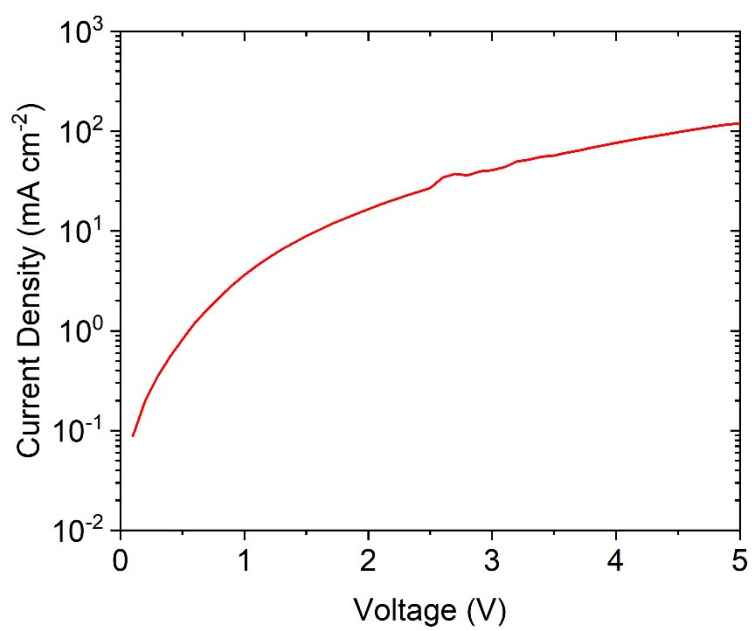
To avoid the effects of oxygen, both as a cause of chemical degradation and as a triplet quencher, samples were placed in a vacuum chamber and evacuated to less than  $2 \times 10^{-5}$  mbar. The sample could be photoexcited, and the photoluminescence measured, through the quartz windows.

Where relevant, devices were driven with a Keithley 2400 source meter.

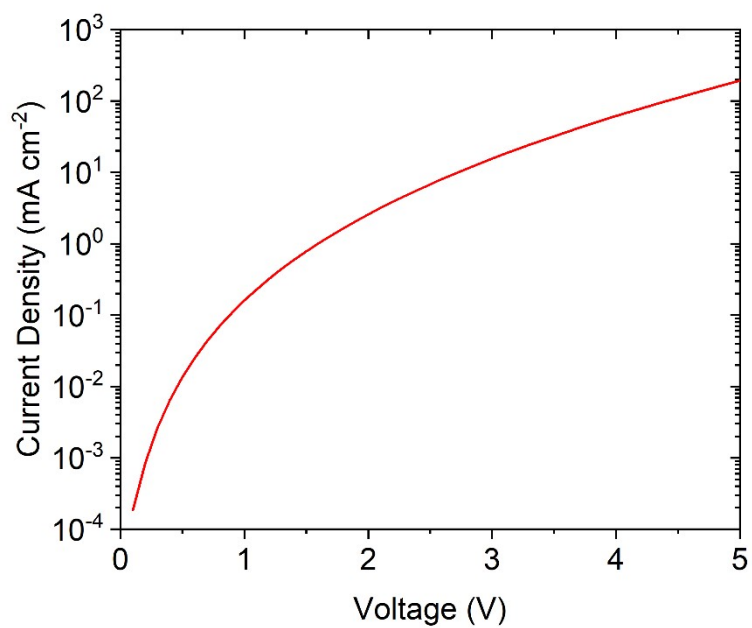
## Supplementary Figures



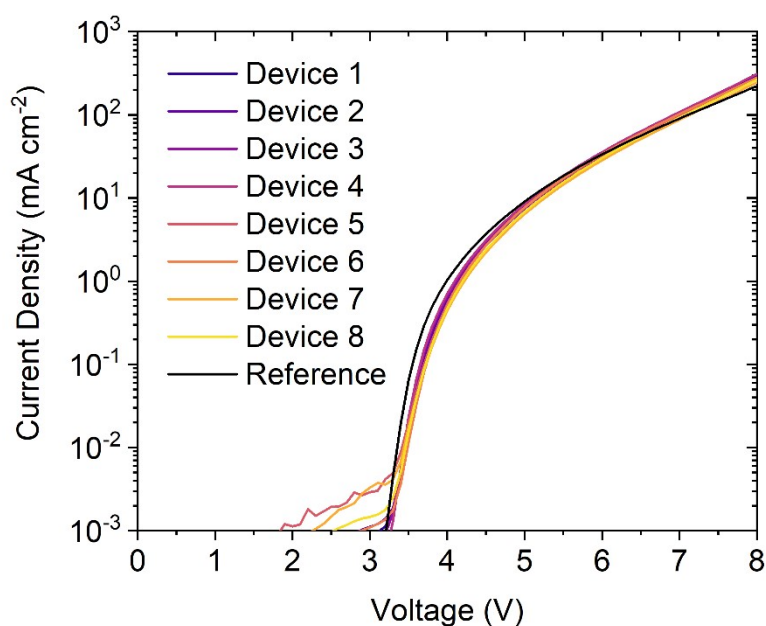
**Figure S1.** Performance characteristics of CMA1 OLEDs. A) Current density-voltage, B) luminance-voltage, C) electroluminescence and D) external quantum efficiency



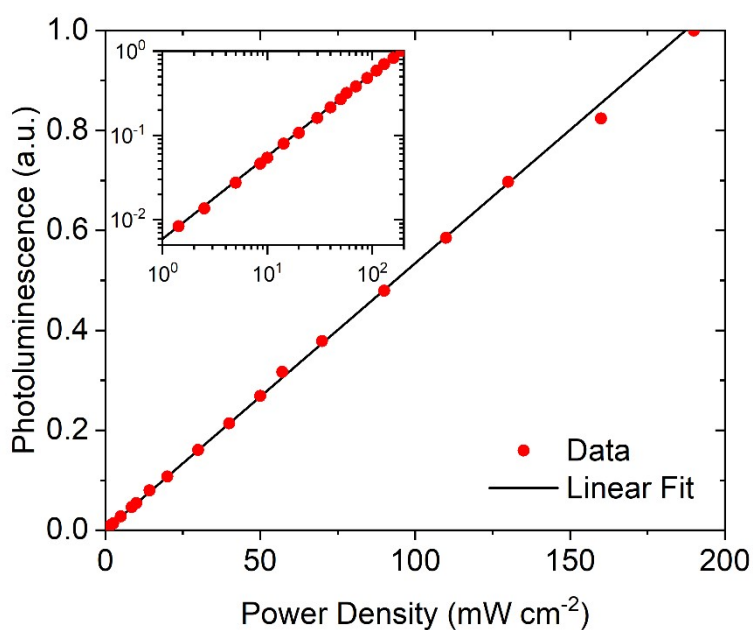
**Figure S2.** Current density-voltage characteristic of electron-only devices



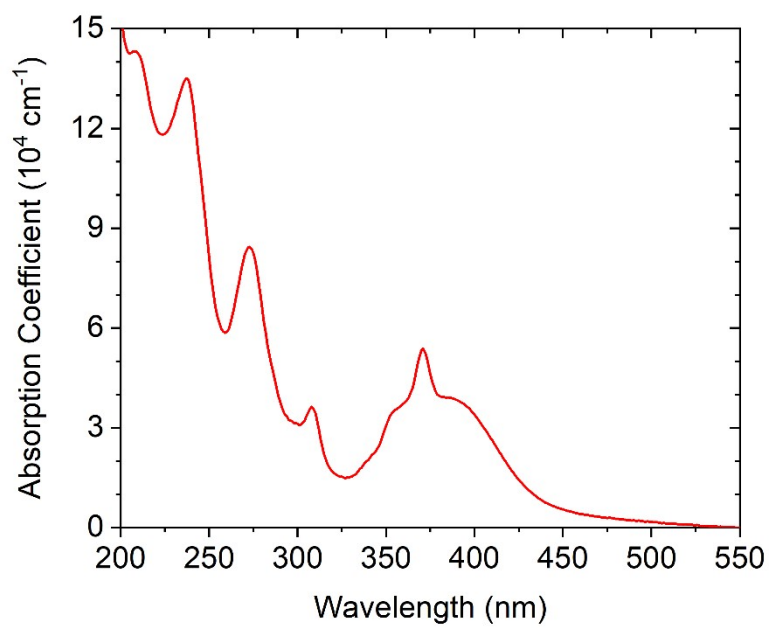
**Figure S3.** Current density-voltage characteristic of hole-only devices



**Figure S4.** Current density-voltage characteristics of a reference CMA1 OLED without DCJTB doping, and of the sensitised devices with 1vol% doping of DCJTB. The device numbers refer to the location of the sensitizer strip with 1 being at the hole-injecting (TAPC:CMA1) interface and 8 at the electron-injecting (CMA1:UGH2) interface.



**Figure S5.** Photoluminescence counts as a function of power density for a CMA1 film excited with a 405 nm laser (inset: the same data on a log-log scale). The linear relationship implies that the exciton density is proportional to the power density.



**Figure S6.** Absorption spectrum of CMA1. Used to calculate the exciton density for a given power density.