Supporting Information

Electrically Driven White Light Emission from Intrinsic Metal–Organic Framework

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**Time Resolve Photoluminescence**

**Table S1:** Time resolved photoluminescence decay lifetime of the free \(\text{H}_4\text{ntca}\) ligand and the MOF.

<table>
<thead>
<tr>
<th>Peak Position (nm)</th>
<th>Decay time ((\tau_1)) (nS)</th>
<th>Decay time ((\tau_2)) (nS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>430 (Free (\text{H}_4\text{ntca}) ligand)</td>
<td>0.98 ± 0.2</td>
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</tr>
<tr>
<td>435 (MOF)</td>
<td>0.60 ± 0.2</td>
<td>--</td>
</tr>
<tr>
<td>507 (MOF)</td>
<td>1.41 ± 0.2</td>
<td>10.87 ± 0.2</td>
</tr>
<tr>
<td>568 (MOF)</td>
<td>2.31 ± 0.2</td>
<td>11.90 ± 0.2</td>
</tr>
<tr>
<td>640 (MOF)</td>
<td>1.28 ± 0.2</td>
<td>10.92 ± 0.2</td>
</tr>
</tbody>
</table>

The extracted photoluminescence decay lifetimes are tabulated in **Table S1**. The obtained lifetime corresponds to a fluorescence emission, and the decay lifetimes are in good agreement with measured lifetimes reported by other groups.\(^1,2\) The PL emission spectrum is the superimposition of different types of transitions that are involved in the composite. The PL peak centered at 435 nm is mainly due to a ligand based emission. The decay lifetime of the free ligand shows that the ligand based decay has a lifetime of less than 1 ns. The peak centered at 507 nm is mainly due to the intermetallic transition and superimposition of the ligand based emission along with an emission centered at 568 nm, which is consistent with the observation of multiple decay times in **Figure 4e**. Because the metal and ligand based emissions have short lifetimes, ~1 ns, the longer decay time could be related to the lifetime of the transition centered at 568 nm. Hence, the emission centered at 568 nm in **Figure 4c** can be related to intersystem charge transfer, which has been shown to have a longer lifetime.\(^3,4\) Similarly, the peaks centered at 568 nm and 640 nm are a mixture of a metal based emission and an emission due to intersystem charge transfer.\(^1\) The errors in the lifetime measurements are limited by the response time of the PMT detector, which was found to be ~200 ps.
Optical Transparency of Graphene/PMMA:

Figure S1 | Transmittance spectrum of graphene and 300 nm PMMA.

A thin PMMA layer was used as a carrier film for transferring graphene which was made thinner (~300 nm) in order to use it as a window layer along with graphene. The PMMA layer provides mechanical strength to the graphene and prevent cracks due to the surface roughness of the MOF layer. It also protects the graphene from atmospheric effects. The transmittance spectrum of graphene/PMMA as shown in Figure S1 indicates a high transmittance of optical photons through the window layer, composed of graphene/PMMA, indicating that a good quality window layer was formed.

Commercial-WLED and MOF-WLED

Figure S2 | Commercial-WLED and MOF-WLED spectrum comparison.
A comparison of MOF-WLED with a commercially available WLED of the same color temperature reveals an important fact that the intense blue emission of the commercial WLED can be replaced by continuous spectrum white light using the MOF as an electroluminescent material. Suitable band alignment is strictly necessary in order to activate the emission due to the different components, which produces multiple bandgaps in the supermolecule.

**Role of H$_2$O Molecules in Emission Process**

![Temperature dependent PL spectra of compound 1 under an excitation at 266 nm.](image)

*Figure S3* | Temperature dependent PL spectra of compound 1 under an excitation at 266 nm.

Temperature dependent PL spectra were obtained in order to study the role of guest molecules in the emission process. The TGA data in *Figure 1b* reveals that the guest water molecules can be completely eliminated at a temperature higher than 88 ºC. Since the emission spectrum remains unchanged at 90 ºC in *Figure S3*, it can be concluded that guest molecules do not take part in the emission process. The emission spectrum changes dramatically at 250 ºC. At this temperature both guest and coordinate water molecules are removed, as shown in *Figure 1b* while the PXRD data in *Figure 1c* suggests that the crystal structure remains intact. Thus, coordinating water molecules play an important role in the emission process by broadening the spectrum. As the coordinate water was removed from compound 1, the metal based emission becomes much higher due to the elimination of O-H vibrational quenching.
In addition to spectral broadening, the coordinate water molecules in compound 1 cause the appearance of an emission centered around 560 nm with a longer lifetime, which is due to a MLCT emission assisted by coordinated water molecules.

**Origine of Sharp Peaks**

![Figure S4](image)

**Figure S4** | PL spectrum of Compound 1 at a higher excitation power (45 µW).

Under a 266 nm UV illumination, compound 1 has a broad emission, which results in a white color spectrum. Strontium (Sr) metal plays an important role in the emission. As marked in **Figure S2**, Sr has several interatomic transitions which belong to the visible range. At a lower pumping power the broad band emission is dominant over the atomic level transitions, which could be the reason for the disappearance of all of the sharp peaks. The sharp peak at around 580 nm appears to be due to charge transfer from metal to ligand energy levels.

**Diffuse Reflectance Spectrum of Compound 1**
**Figure S5** | Diffuse reflectance spectrum of compound 1. The noise due to the lamp switch is denoted with a circle.

**Electrochemical stability of the device:**

**Figure S6** | Electrochemical stability of the device.

The electrochemical stability of the device was evaluated by current-voltage (I-V) measurements of the device before and after the application of a constant voltage of 10 V for 6 hours. The similar nature of I-V confirms that the device has a high electrochemical stability.
CVD grown graphene was used the window layer of the WLED. The Raman spectrum of the graphene layer on SiO$_2$ is shown in Figure S7. The intensity ratio, $I_G/I_{2D}$ of the 2D to the G peak was found to be <1 and again, the 2D peak profile is Lorentzian in nature, which confirms that a single layer of graphene formed.\textsuperscript{6,7} Again, the absence of a D peak at ~ 1323 cm$^{-1}$ indicates an almost defect-free high quality graphene layer.\textsuperscript{6,7}

**ZnO Nanoparticles (ZnO NPs):**

![Figure S8](image)

**Figure S8** | ZnO nanoparticles (ZnO NPs). a, Absorption and emission spectrum of ZnO NPs. The PL emission was studied under the excitation of 266 nm laser. b, Scanning Electron Microscopy image of ZnO NPs coated on Si/SiO$_2$ substrate.

The hydrothermally synthesized ZnO NPs were coated on a quartz substrate for measuring its UV-vis absorption and photoluminescence (PL) emission. The absorption and
emission spectra are shown in Figure S8a. The scanning electron microscopy (SEM) image of the ZnO NP layer is shown in Figure S8b. The average size of the ZnO NP was determined to be ~6 - 8 nm.

References:


