Supporting Information

for

Photo-enhanced lithium-ion batteries using metal-organic framework

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Experimental Section

Material synthesis: First, a 150 ml solution of 480 mg (~ 2.61 mmol) copper (II) acetate and 150 mg (~ 0.51 mmol) sodium dodecyl sulfate was prepared in a 500 ml conical flask. Then, 150 ml NaOH (150 mg, 3.75 mmol) was added to the above solution, followed by addition of 300 mg (~ 1.74 mmol) tetrahydroxy-1,4-quinone powder. The solution was sonicated for 30 min at 50 °C and then left overnight. The final precipitate was collected and washed with water and ethanol in sequence and ultrasonicated in an ice bath for 30 minutes. After that, the sample was collected and dried in a vacuum oven at 80 °C.¹

Ph-LIB assembly: First, an electrode solution containing Cu-HHB, rGO, and PVDF in a wt% ratio of 85:10:5 was prepared in NMP solvent using sonication and mixing processes. Thereafter, the material was drop cast on CF current collectors followed by drying at 120 °C in a vacuum oven. Electrodes as obtained from this process we used with Li metal foil counter electrodes and 5M LiTFSI in EC/PC (v/v = 1/1) to assemble optical coin cells, with an optical window of ~ 7 mm in diameter by following the referenced procedure.²

Electrochemical measurement: The electrochemical tests of the Ph-LIB were measured by using Biologic VMP-3 and BCS potentiostats. CV at different scan rates $(0.1 - 1.0 \text{ mV s}^{-1})$ and GDC at different specific currents $(100 - 2000 \text{ mA g}^{-1})$ were tested in the dark and 1 sun illuminated conditions. The EIS tests of the Ph-LIB were recorded in the frequency range 10 mHz to 100 kHz at 10 mV amplitude both in the dark and 1 sun illuminated (LSH-7320 LED Solar Simulator) conditions.

Fabrication of PDs and electrical measurements: The electrical photoresponse of Cu-HHB was studied by fabricating metal-semiconductor-metal PD. In this process, Au/chromium (Cr) (40/10 nm) interdigitated electrodes (IDEs) on a Si3N4/Si wafer using UV lithography and subsequently drop-cast Cu-HHB on the IDEs. Current-voltage tests of the PD were measured by sweeping the voltage from 0 to +2 V both in dark and light illuminated conditions. Moreover, the current time of the PD was tested under alternating dark and illuminated conditions in the presence of an external bias voltage of 2 V. On the other hand, the stack type FTO/rGO/Cu-HHB/Ag PD was fabricated by layer-by-layer coating of rGO and Cu-HHB on the transparent FTO coated glass substrate. Then, the current-time test of the stacked PD was studied in alternating dark and illuminated conditions in the absence of bias voltage.



Figure S1. Energy band diagram of the photo-cathode.



Figure S2. High magnification SEM image of the Cu-HHB MOF.



Figure S3. High-resolution spectra in the (a) C 1s and (b) O1s regions of Cu-HHB.



Figure S4. (*a*,*b*) *CV* profiles at 0.1 mV s⁻¹ and 0.5 mV s⁻¹ in dark and 1 sun illumination.



Figure S5. Capacitive contribution calculations to charge storage at 1.0 mV s^{-1} in the (a) dark and (b) 1 sun illumination.



Figure S6. (*a-c*) *GDC* profiles at 100 mA g^{-1} , 500 mA g^{-1} and 1000 mA g^{-1} in the dark and 1 sun illuminated conditions.

References

- 1. Z. Wang, G. Wang, H. Qi, M. Wang, M. Wang, et al., Chem. Sci., 2020, 11, 7665-7671.
- 2. B. D. Boruah, B. Wen, and M. D. Volder, Nano Lett., 2021, 21, 3527-3532.