# Electronic Supplementary Information (ESI)

# An Asymmetric Supercapacitor Based on a Non-Calcined 3D Pillared Cobalt(II) Metal-Organic Framework with Long Cyclic Stability

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**Materials and physical techniques.** All starting materials for the synthesis were purchased from commercial providers and used without further purification. FT-IR spectra were recorded using a Nicolet Fourier Transform IR 100 spectrometer in the 400-4000 $cm^{-1}$ range using the KBr disk technique. The thermal behavior of the samples was analyzed by a PL-STA 1500 apparatus working under a nitrogen atmosphere and at the heating rate of 10 °C $min^{-1}$. Powder X-ray diffraction (PXRD) measurements were performed using a Bruker AXS model D8 advanced with monochromated Cu-Kα (λ=1.54056 A) radiation. The $N_2$ adsorption/desorption isotherms were measured at 77 K using a Micromeritics ASAP 2020 analyzer. The specific surface area was calculated by the Brunauer-Emmett-Teller (BET) method. The inductively coupled plasma (ICP) analysis was performed on a Varian ICP-OES VISTA-PRO CCD instrument. A CHNS Thermo Scientific Flash 2000 elemental analyzer was used to analyze elemental contributions to the samples. Finally, melting points were measured on an Electrothermal 9100 apparatus.

**X-ray diffraction structure determinations**: Single crystals coated with viscous hydrocarbon oil were mounted on loops. Data of Co(II)-TMU-63 were obtained at −173 °C (100 K) on the MX1 beamline at the Australian Synchrotron operating at 17.4 keV (λ = 0.7109 Å) with data collection conducted using Blu-Ice control software.[S1] The diffraction data were processed, reduced and corrected with the XDS software suite.[S2] The structures were solved by conventional methods and refined by full-matrix least-squares on all $F^2$ data using SHELX2014,[S3] in conjunction with the X-Seed[S4] or Olex2[S5] graphical user interface. All hydrogen atoms were placed in calculated positions using the riding model. Crystal data and refinement details are given in Table S1. CCDC-1944674 contains the supplementary crystallographic data for this

paper. These data can be obtained free of charge from The Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

**MOF activation method.** Under vacuum, each sample of Co(II)-TMU-63 was heated at 140 ℃ in an oven for 48 h. When all DMF moieties are removed, the structure experiences no damage or impairment.

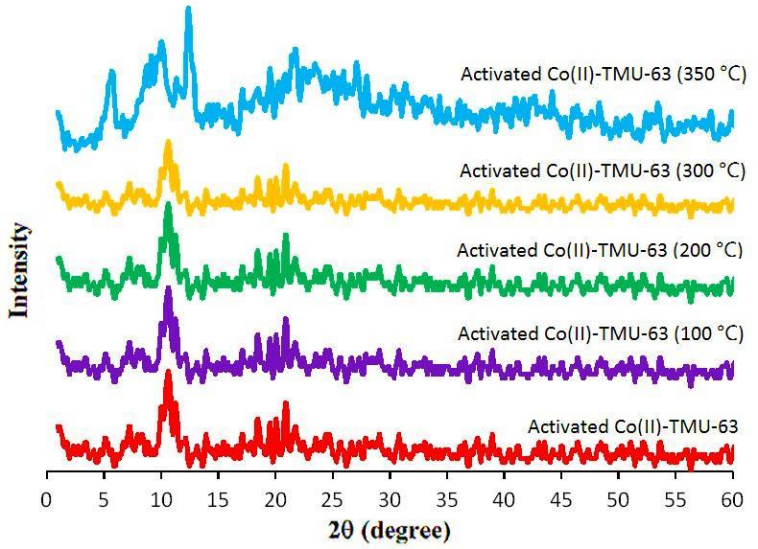


**Figure S1.** PXRD patterns of Co(II)-TMU-63 at different temperatures.

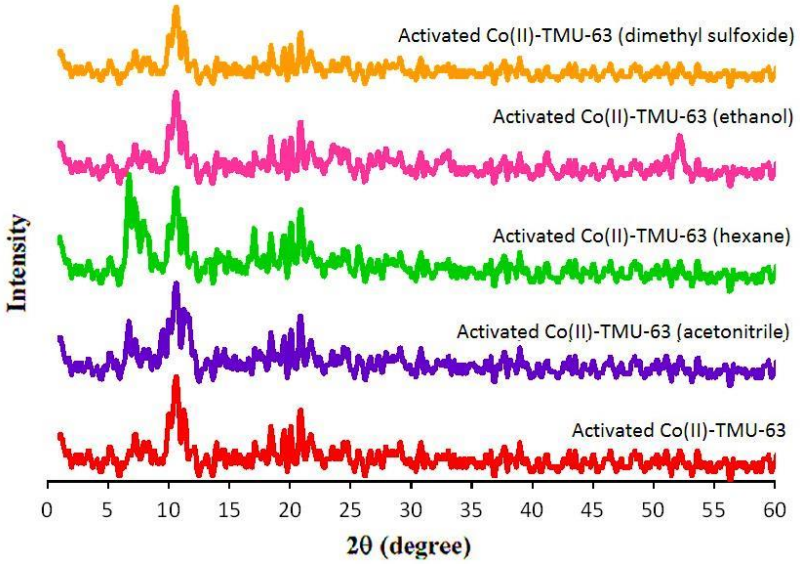


**Figure S2.** PXRD patterns of Co(II)-TMU-63 after refluxing for 24 h in different solvents.

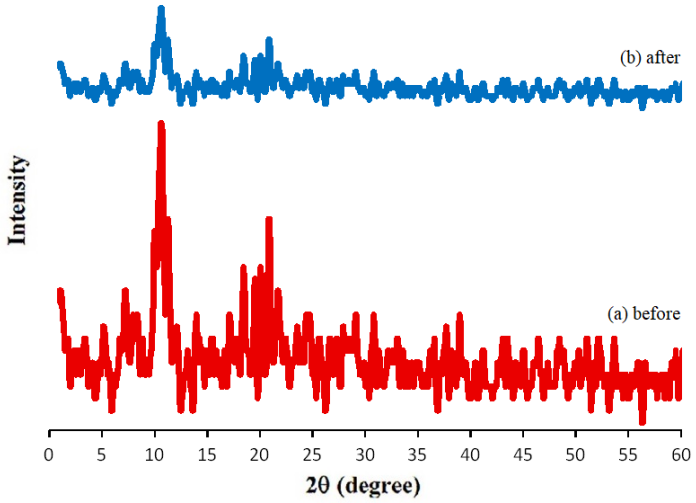


**Figure S3.** PXRD patterns of Co(II)-TMU-63 before and after cyclic stability test.

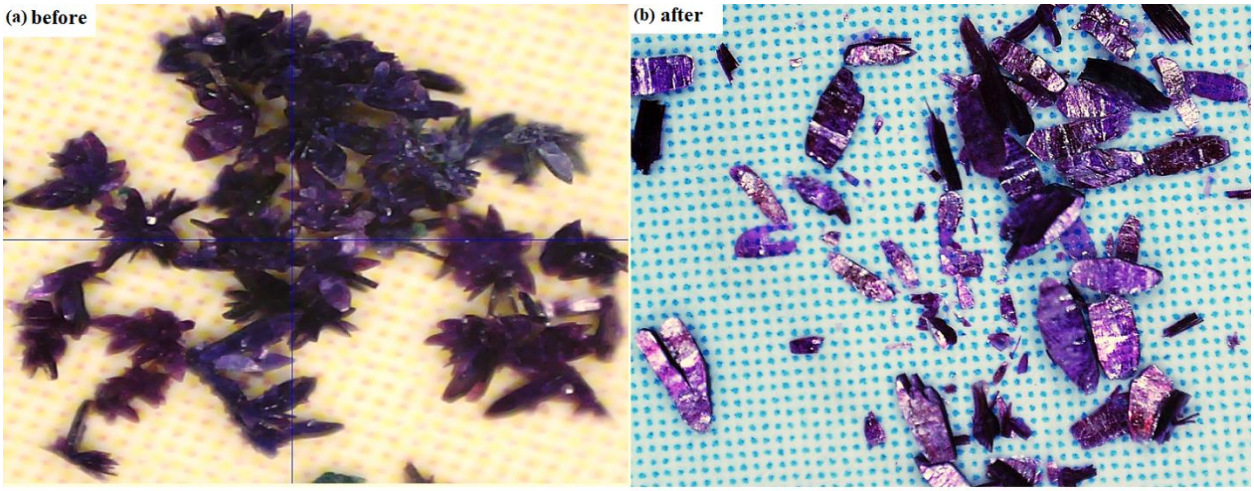


**Figure S4.** Light microscope image of solvated bulk sample of the Co(II)-TMU-63 crystals (a) before and (b) after cyclic stability test.

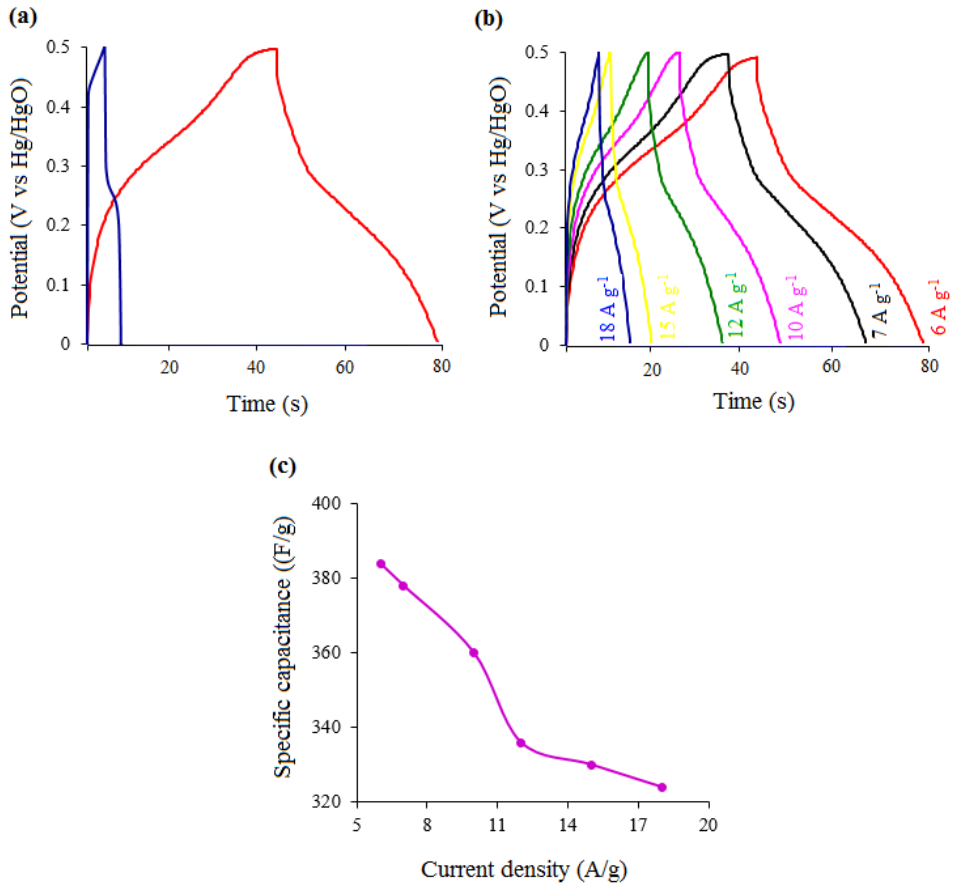


**Figure S5.** Charge-discharge curves of the Co(II)-TMU-63 electrode material: (a) at 6 A $g^{-1}$ current density and (b) at various current densities from 6 to 18 A $g^{-1}$. (c) Relationship of the specific capacitance and current densities from 6 to 18 A $g^{-1}$.

**Table S1.** Crystal data and structure refinement for Co(II)-TMU-63.

| | |
|---|---|
| Identification code | Co(II)-TMU-63 |
| Empirical formula | $C_{20}H_{21}Co_{1.50}N_4O_8$ |
| Formula weight | 533.80 |
| Temperature/K | 173(2) |
| Crystal system | monoclinic |
| Space group | C 2/c |
| a/Å | 31.351(6) |
| b/Å | 9.5960(19) |
| c/Å | 18.028(4) |
| α/° | 90 |
| β/° | 118.68(3) |
| γ/° | 90 |
| Volume/$Å^3$ | 4758(2) |
| Z | 8 |
| $\rho_{calc}g/cm^3$ | 1.490 |
| F(000) | 2188 |
| Crystal size/$mm^3$ | 0.240 |
| Radiation | Mo\a (λ = 0.71073) |
| 2Θ range for data collection/° | 1.481 to 24.99 |
| Index ranges | -37 ≤ h ≤ 37, -11 ≤ k ≤ 11, -21 ≤ l ≤ 21 |
| Reflections collected | 27245 |
| Independent reflections | 11818 [$R_{int}$ = 0.0722, $R_{sigma}$ = 0.1427] |
| Data/restraints/parameters | 4186/9/283 |
| Goodness-of-fit on $F^2$ | 1.062 |
| Final R indexes [I>=2σ (I)] | $R_1$ = 0.0773, $wR_2$ = 0.1978 |
| Final R indexes [all data] | $R_1$ = 0.0707, $wR_2$ = 0.2048 |
| Largest diff. peak/hole / e $Å^{-3}$ | 1.923/-1.461 |

## Reference


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