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## **1. Experimental details.**

### **1.1 Synthesis of fullerene C<sub>70</sub> cubes (FC).**

Fullerene C<sub>70</sub> cubes (FC) were synthesized by the ultrasound-assisted liquid-liquid interfacial precipitation (ULLIP) method at 25 °C. Fullerene C<sub>70</sub> solution in mesitylene (1 mg mL<sup>-1</sup>) was prepared by dissolving pristine C<sub>70</sub> powder (30 mg: 99 % pure, MTR Ltd., USA) in mesitylene (30 mL). Bath sonication was applied for 1 h to completely dissolve the pristine C<sub>70</sub> powder. In a typical crystallization, C<sub>70</sub> solution (1 mL) was placed in a thoroughly cleaned and dried glass bottle (13.5 mL) and stored in an incubator at 25 °C for 30 min to attain the equilibrium temperature. Tertiary butyl alcohol (TBA: 5 mL), which had also been stored at 25 °C, was added slowly to the C<sub>70</sub> solution so that a clear liquid-liquid interface was formed. The above mixture was incubated without disturbance at 25 °C for 3 h. Gentle shaking by hand was then applied followed by bath sonication for 30 s. The resulting mixture was again stored at 25 °C in an incubator for 48 h to obtain fullerene C<sub>70</sub> cubes (referred to FC hereafter). The as-prepared FC in the mother liquor was stirred (300 rpm) for 72 h at 75 °C to obtain mesoporous crystalline fullerene C<sub>70</sub> cubes (referred to as MCFC). For the large scale synthesis of MCFC, FC in the mother liquor was stirred (300 rpm) for 72 h at 75 °C then separated by centrifugation, and the obtained solid product was dried at 80 °C under reduced pressure for 24 h. MCFC was heat-treated at 220 °C under reduced pressure for 24 h with the product obtained being referred to as MCFC<sub>220</sub>. For a comparison of their structures and properties, FC was also heat-treated at 220 °C under reduced pressure for 24 h with the product obtained being referred to as FC<sub>220</sub>.

### **1.2 Characterizations.**

Scanning electron microscopy (SEM) images of FC, MCFC, and MCFC<sub>220</sub> were obtained using a Hitachi Model S-4800 field effect scanning electron microscope (FE-SEM) operating at an accelerating voltage of 10 kV. SEM samples were prepared by dropping suspensions of FC, MCFC, and MCFC<sub>220</sub> onto passivated silicon substrates followed by drying at 80 °C. All the samples were coated with platinum (~ 2 nm) by sputtering using a Hitachi S-2030 ion coater. Transmission electron microscopy (TEM) images (TEM and HR-TEM), and selected area electron diffraction (SAED) patterns were obtained using a transmission electron microscope (JEOL Model JEM-2100F operated at 200 kV). TEM samples were prepared by dropping suspensions of FC, MCFC, and MCFC<sub>220</sub> onto standard carbon-coated copper grids. TEM samples were dried at 80 °C for 24 h



under reduced pressure prior to TEM measurements. Surface functional groups were identified by Fourier transform infrared (FT-IR) spectroscopy. FT-IR spectra of pristine fullerene C<sub>70</sub> and FC, MCFC, and MCFC\_220 were recorded using a smart-detector-equipped Nicolet 4700 FT-IR instrument, Thermo Electron Corporation. Thermogravimetric analyses of FC and MCFC were performed using a SII Instrument (Model Exstar 600) with samples under an argon gas atmosphere at a heating rate of 10 °C min<sup>-1</sup>. Raman scattering spectra were collected using a Jobin-Yvon T64000 Raman spectrometer. Raman samples were prepared on a clean glass slide followed by drying at 80 °C on a dry bath. Excitation was performed using a green laser of wavelength 514.5 nm at 0.02 mW power for 20 s. Powder X-ray diffraction (XRD) patterns were recorded at room temperature on a Rigaku RINT2000 diffractometer with Cu-K $\alpha$  radiation ( $\lambda$  = 0.1541 nm). Nitrogen adsorption-desorption isotherms were recorded on an automatic adsorption instrument (Quantachrome Instruments, Autosorb-iQ2, USA) at liquid nitrogen temperature 77.35 K. For each measurement, an aliquot (20 mg) of the respective sample was degassed for 24 h at 100 °C prior to the measurements.

## 1.2 Electrochemical tests.

Electrochemical performances of pristine C<sub>70</sub>, FC, MCFC, and MCFC\_220 were evaluated by cyclic voltammetry (CV) using a Electrochemical Analyzer, Model 850D, ALS/CH instruments in the potential range from 0 to 0.8 V (vs. Ag/AgCl) using 1.0 M H<sub>2</sub>SO<sub>4</sub> aqueous solution as electrolyte. Platinum wire was used as the counter electrode with Ag/AgCl as the reference electrode. A clean bare glassy carbon electrode (GCE) was used as a working electrode. The working electrode was modified by applying a dispersion of the samples. Typically, pristine C<sub>70</sub> (1 mg) was suspended in 4:1 v/v water-ethanol mixture (1 mL) followed by sonication for 10 min. The resulting dispersion (3  $\mu$ L) was placed on the GCE surface and dried at 60 °C for 1h. Nafion solution (0.5 % w/v, 3 $\mu$ L) was then added on the modified working electrode surface as a binder followed by drying at 60 °C for 12 h. Working electrodes for FC, MCFC and MCFC\_220 were each prepared using the same method.

From CV data, gravimetric specific capacitances ( $C_s$ ) of the electrode material were calculated from equation (1):

$$C_s = \frac{\int I dv}{m \times v \times \Delta V} \quad (1)$$

where  $I$  (A),  $v$  ( $\text{mV s}^{-1}$ ),  $\Delta V$  (V) and  $m$  (g) represents current, scan rate, operating voltage and mass of the active electrode material, respectively.

Volumetric capacitance was also calculated (three-electrode system) as

$$C_v (\text{F cm}^{-3}) = \rho \times C_s \quad (2)$$

where  $\rho$  is the compaction density of the electrode material and  $C_s$  is the gravimetric specific capacitance ( $\text{F g}^{-1}$ ). Compaction density of the electrode material,  $\rho$  was calculated as

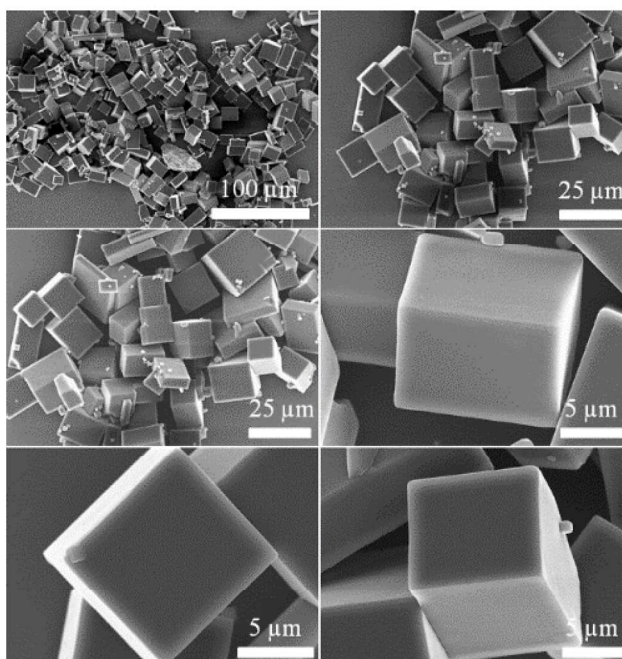
$$\rho = (V_{\text{total}} + 1/\rho_T)^{-1} \quad (3)$$

where  $V_{\text{total}}$  is the total pore volume estimated from the nitrogen sorption isotherm (@77 K),  $\rho_T$  is the true density of the material determined by helium density.

### 1.3 Photoluminescence (PL) and Time-resolved PL (TRPL).

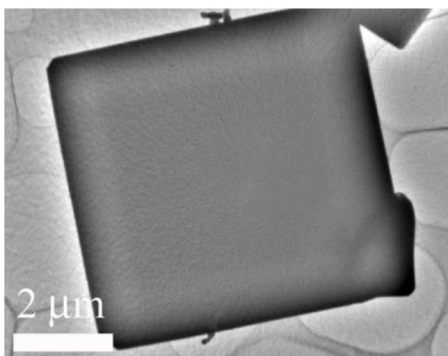
Photoluminescence (PL) spectra of FC, MCFC and MCFC\_220 were measured at room temperature (25 °C) in air using a spectrometer (Acton Research SpectraPro 300i) equipped with a liquid-nitrogen-cooled CCD detector (Roper Scientific Spec10-100BLT/LN). PL spectra of pC<sub>70</sub> and FC\_220 were also recorded for comparison. Samples (0.5 mg mL<sup>-1</sup>) were cast on a silicon substrate and photoexcited using the 442.1 nm line of a He-Cd laser under ambient conditions. All PL spectra have been corrected for the wavelength-dependent sensitivity of the photodetection system. TRPL curves were obtained by means of a time-correlated single photon counting method using the second harmonic generation of 800-nm-line of a Ti:sapphire laser (MIRA 9300, Coherent) pumped by a solid-state green laser (Verdi, Coherent).

## 2. SEM images of fullerene C<sub>70</sub> cubes (FC)



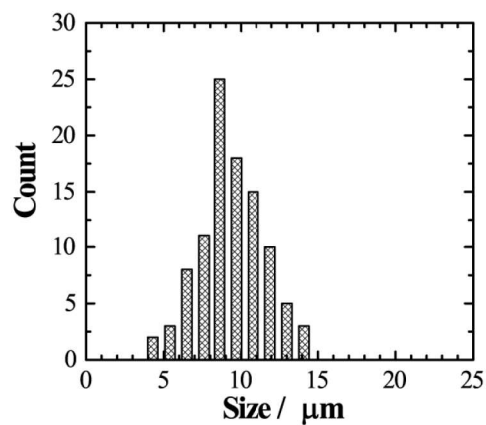
**Figure S1:** SEM images of FC produced at the interface between TBA and C<sub>70</sub> solution in mesitylene at 25 °C. Volume ratio of mesitylene and TBA was fixed at 1:5 and pre-incubation time is 3 h.

## 3. TEM images of FC



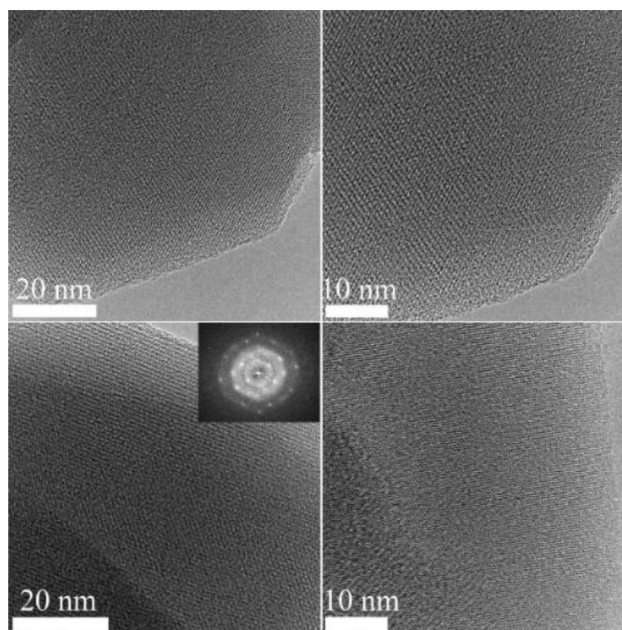
**Figure S2:** TEM image of FC.

#### 4. Histogram of size distribution of FC



**Figure S3:** Histogram of the size distribution of FC obtained at mesitylene-TBA interface with pre-incubation time of 3 h.

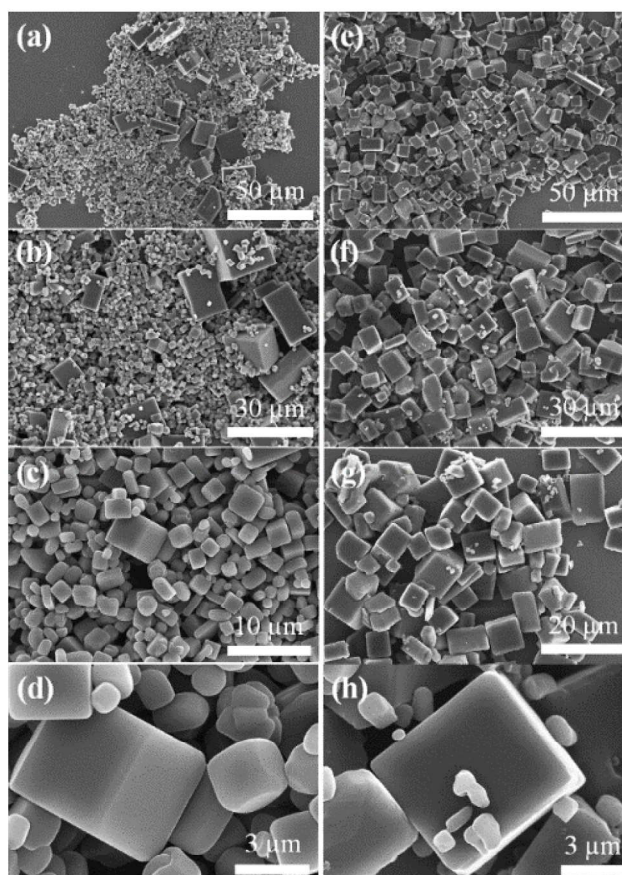
#### 5. HR-TEM images of FC



**Figure S4:** HR-TEM images of FC.

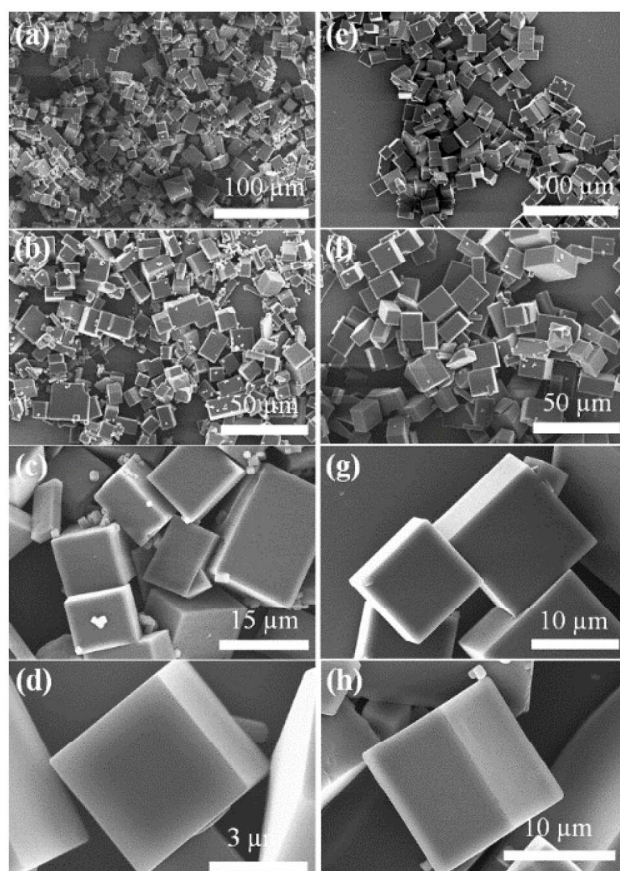


6. SEM images of FC at different volume ratios of solvent and anti-solvent



**Figure S5:** SEM images of FC produced at different mixing ratios of mesitylene and TBA: (a-d) 1:1 and (e-h) 1:3.

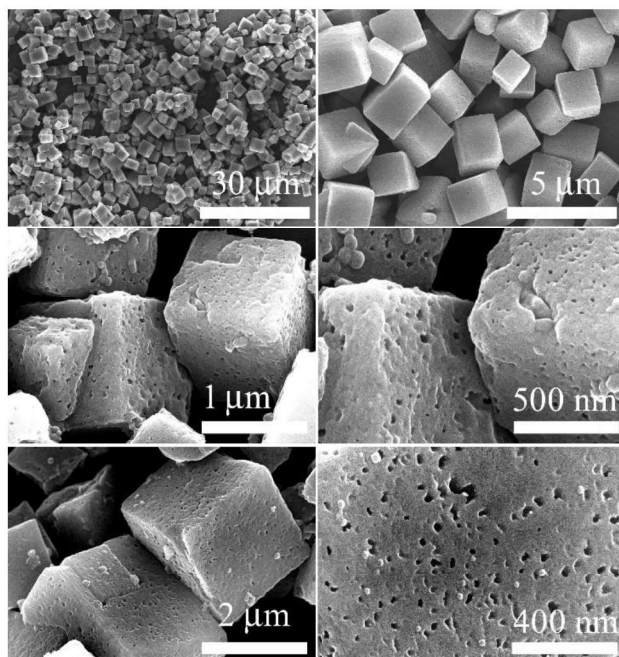
## 7. SEM images of FC at different pre-incubation time



**Figure S6:** SEM images of FC at different pre-incubation times: (a-d) 1 h, and (e-h) 3 h.

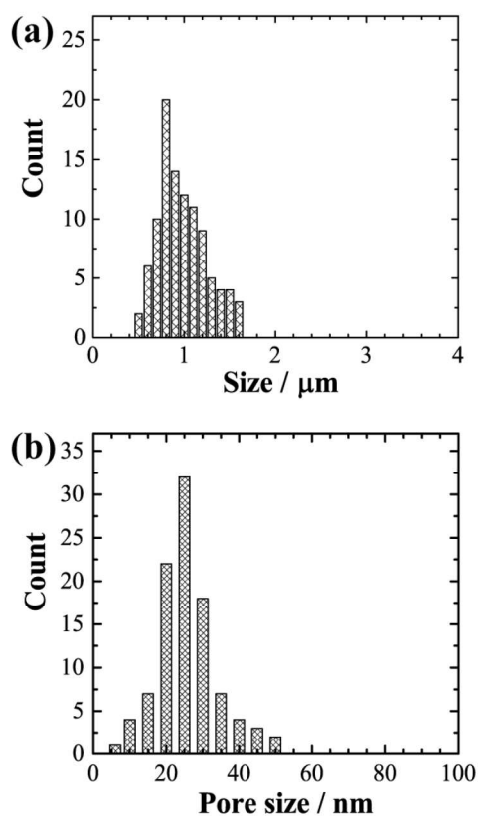


## 8. Additional SEM images of MCFC



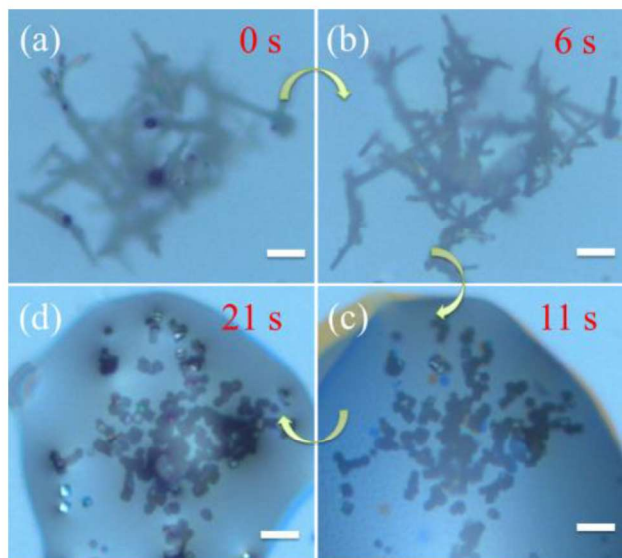
**Figure S7:** Additional SEM images of MCFC.

## 9. Histograms of size distribution of MCFC and pore size distribution



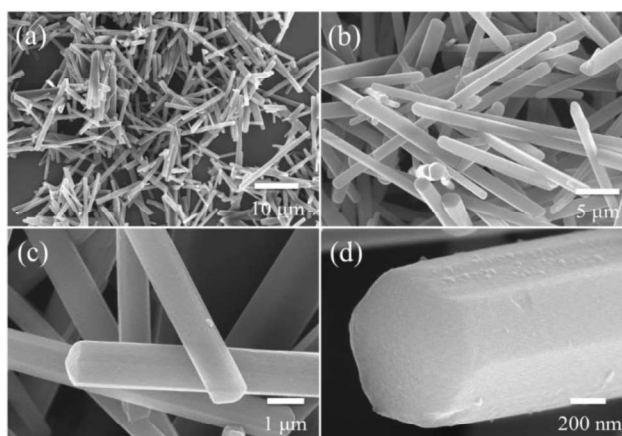
**Figure S8:** (a) Histograms of size distributions of MCFC, and (b) pore size distributions of MCFC based on SEM observations.

#### 10. Optical microscopic images of FNR to MCFC transformations



**Figure S9:** Optical microscopic observation of FNR to MCFC transformation with time. Scale bars are 2  $\mu\text{m}$ .

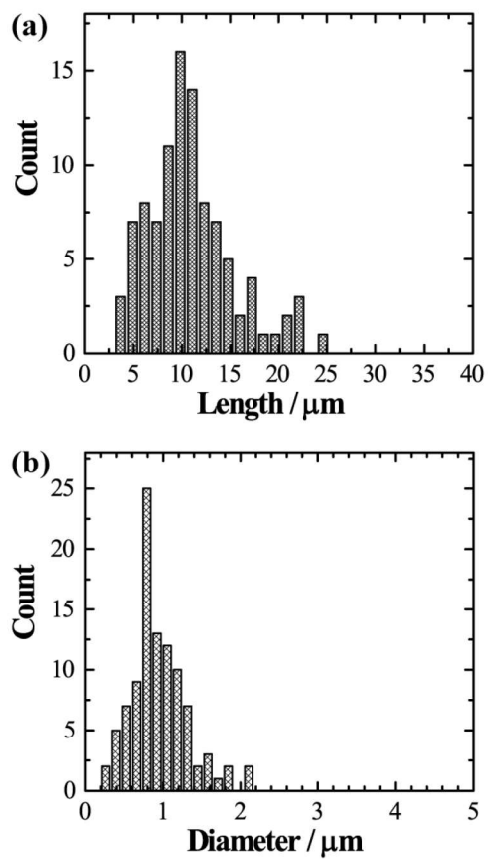
#### 11. SEM images of FNR



**Figure S10:** Typical SEM images of the FNR.

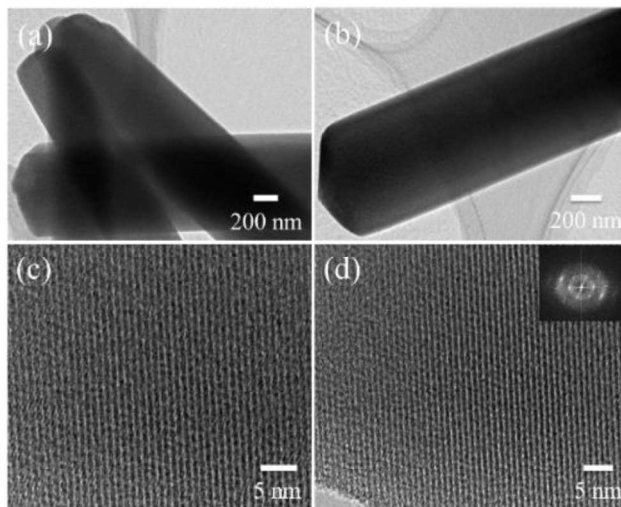


## 12. Histograms of length and diameter distribution of FNR



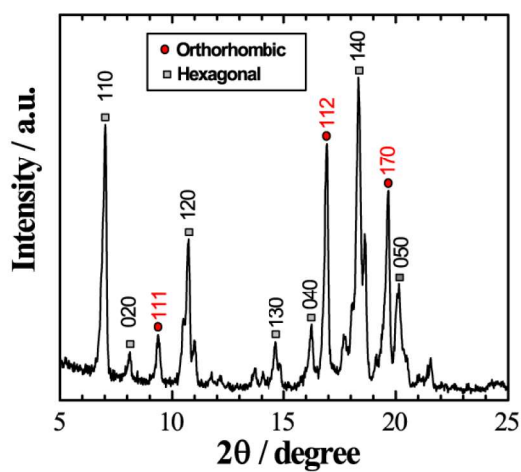
**Figure S11:** (a) Histogram of length distribution of FNR, and (b) histogram of diameter distribution of the FNR.

### 13. TEM and HR-TEM images of FNR



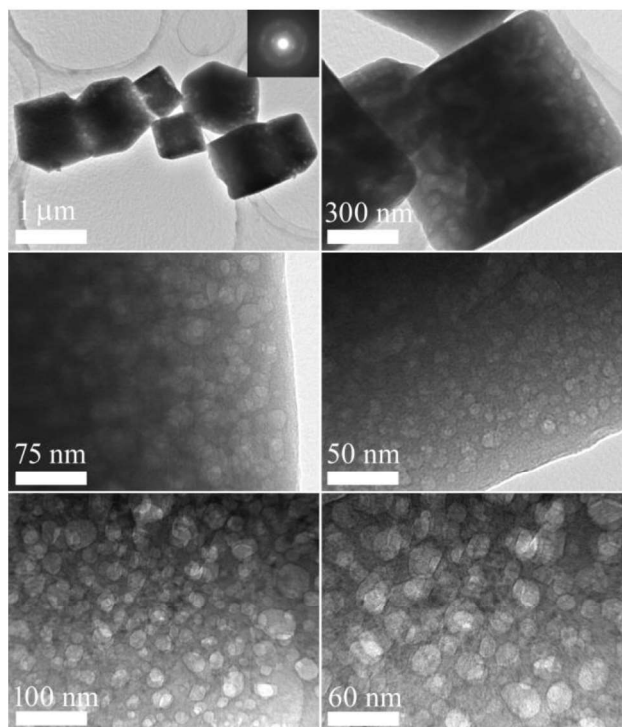
**Figure S12:** (a-b) TEM images and (c-d) HR-TEM images of the FNR.

### 14. Powder XRD pattern of FNR



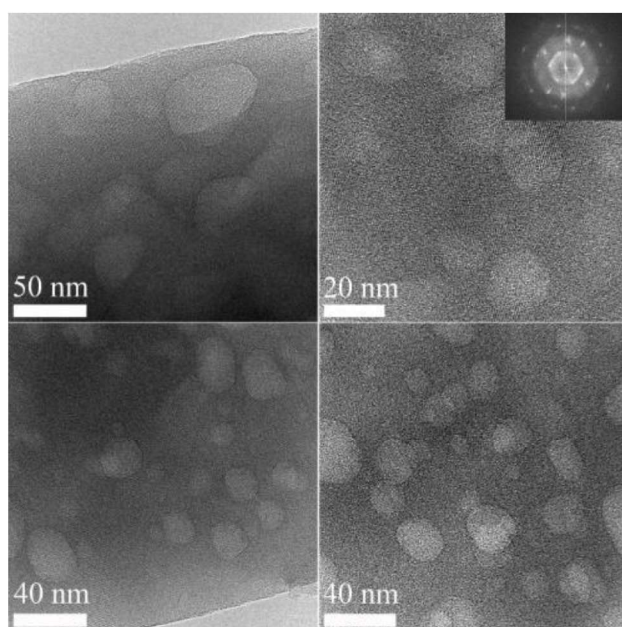
**Figure S13:** Powder XRD pattern of the FNR.

### 15. Additional TEM images of MCFC



**Figure S14:** Additional TEM images of MCFC.

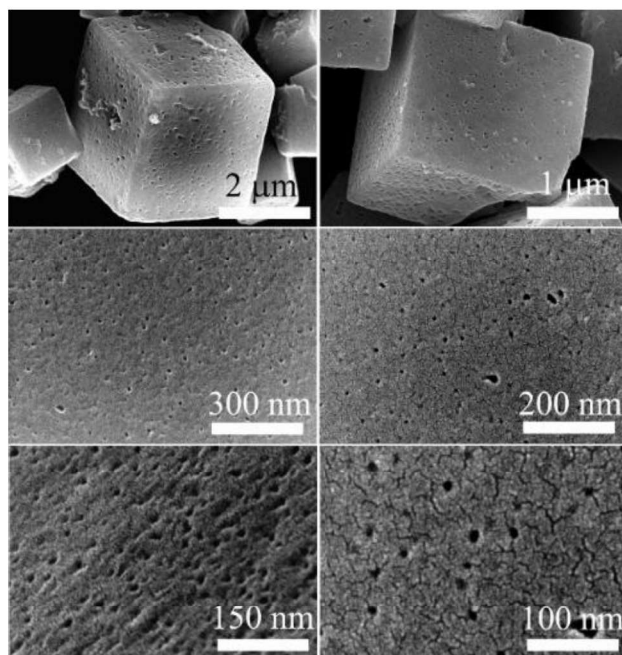
### 16. Additional HR-TEM images of MCFC



**Figure S15:** Additional HR-TEM images of MCFC.

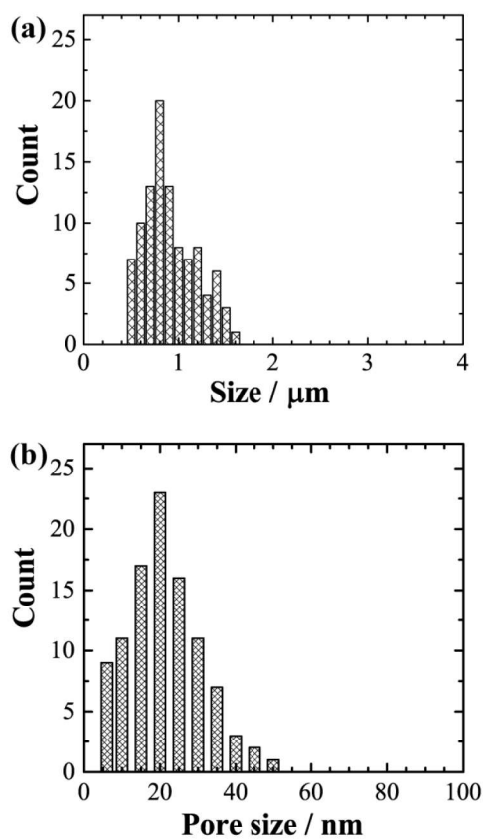


## 17. SEM images of MCFC\_220



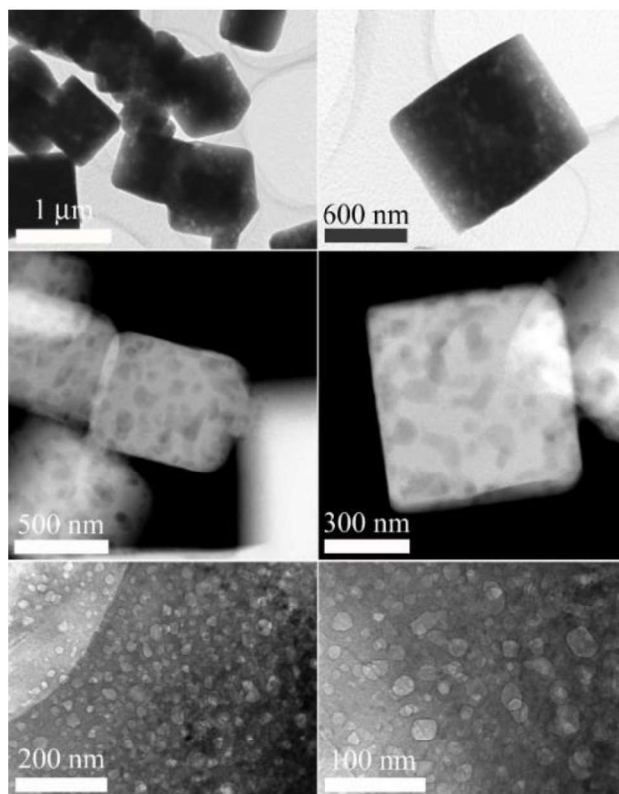
**Figure S16:** SEM and HR-SEM images of MCFC\_220.

## 18. Histograms of size distribution of MCFC\_220 and pore size distribution



**Figure S17:** (a) Histograms of size distribution of MCFC\_220, and (b) pore size distributions of MCFC\_220 determined from HR-SEM image.

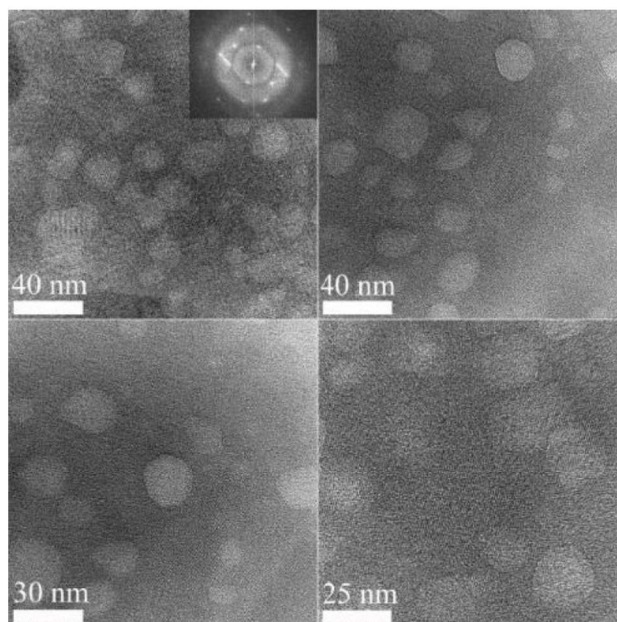
## 19. TEM images of MCFC\_220



**Figure S18:** TEM images of MCFC\_220.

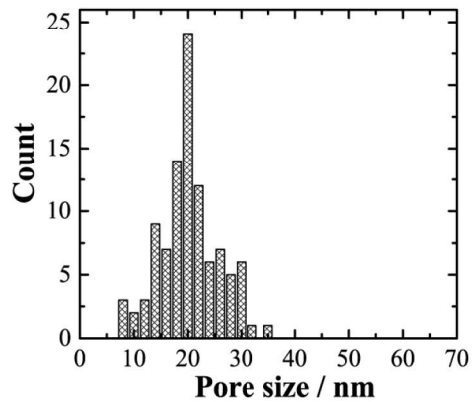


## 20. HR-TEM images of MCFC\_220



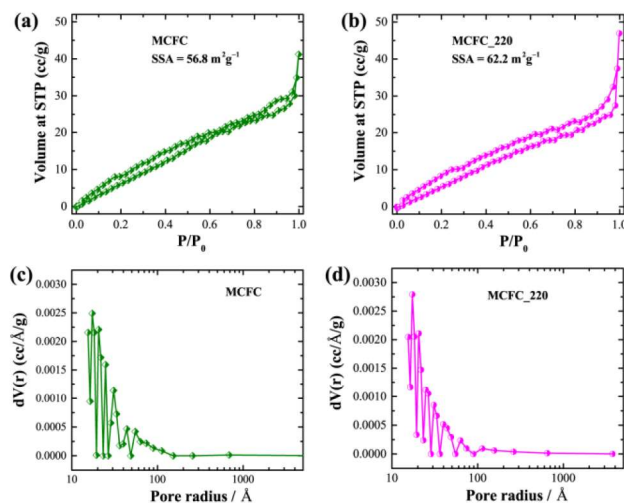
**Figure S19:** HR-TEM images of MCFC\_220.

## 21. Pore size distribution of MCFC\_220 from HR-TEM



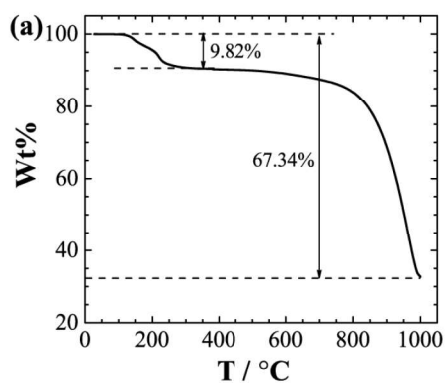
**Figure S20:** Histogram of pore size distribution of MCFC\_220 based on TEM observation.

## 22. Nitrogen adsorption-desorption isotherms



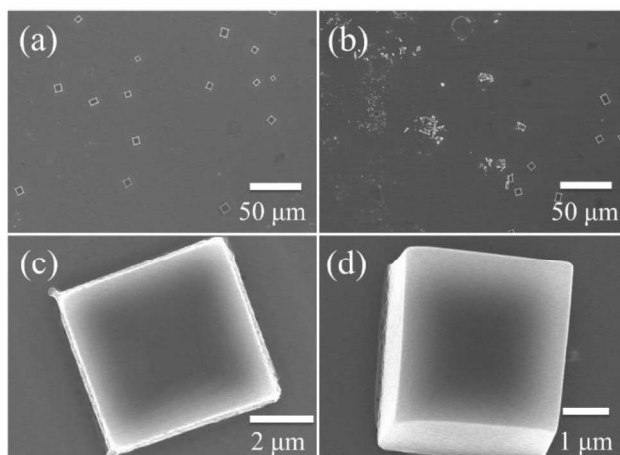
**Figure S21:** (a,b) Nitrogen adsorption/desorption isotherms, and (c,d) corresponding pore size distributions for MCFC, and MCFC\_220.

## 23. Thermogravimetric Analysis (TGA) of MCFC



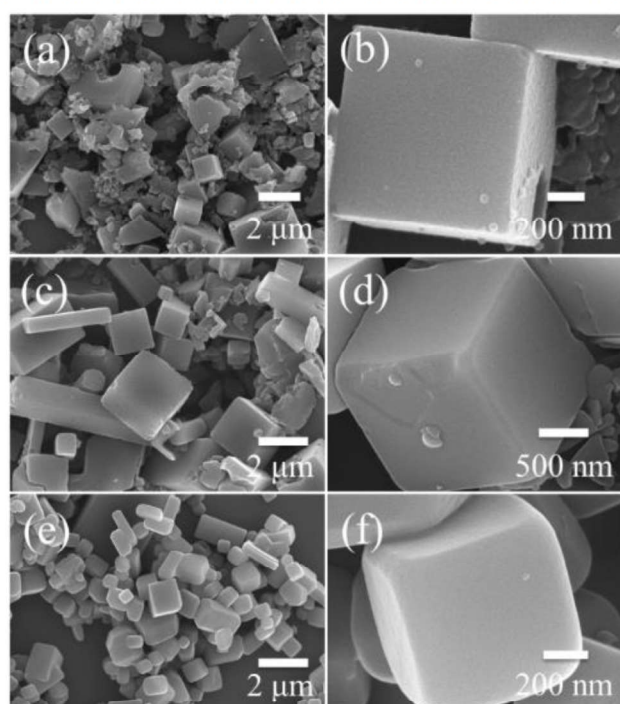
**Figure S22:** TG curve of MCFC under argon atmosphere. The measurement was performed at a heating rate of  $10 \text{ }^\circ\text{C min}^{-1}$ .

#### 24. SEM images of FC obtained by drop casting method



**Figure S23:** SEM images of FC obtained using a drop casting method.  $C_{70}$  solution in mesitylene ( $1 \text{ mg mL}^{-1}$ ) was drop cast on a silicon wafer and dried at  $80^\circ\text{C}$ .

#### 25. SEM images of products obtained from other solvent-antisolvent combinations



**Figure S24:** Products obtained by heating  $C_{70}$  cubes (produced from other solvent-antisolvent combinations) at  $75^\circ\text{C}$  followed by drying on silicon wafer at  $80^\circ\text{C}$ ; (a,b) FC produced from mesitylene-isopropanol, (c,d) mesitylene-ethanol, and (e,f) mesitylene-methanol, respectively.

## 26. Raman scattering spectra

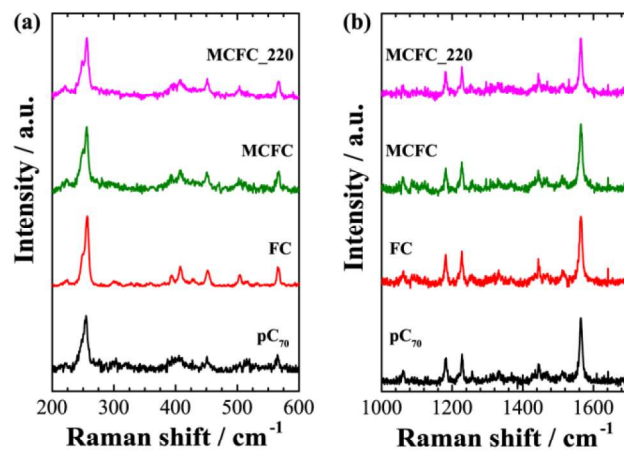
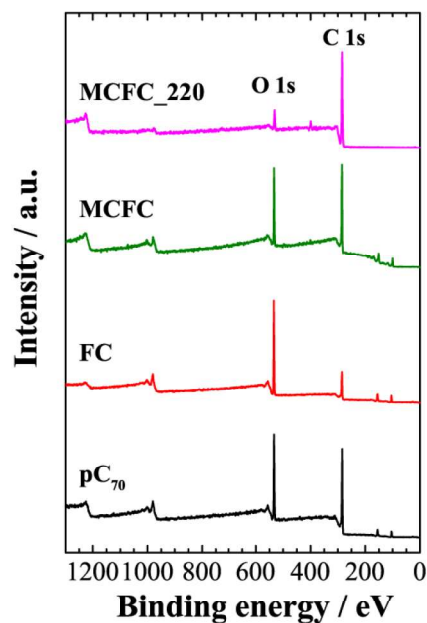
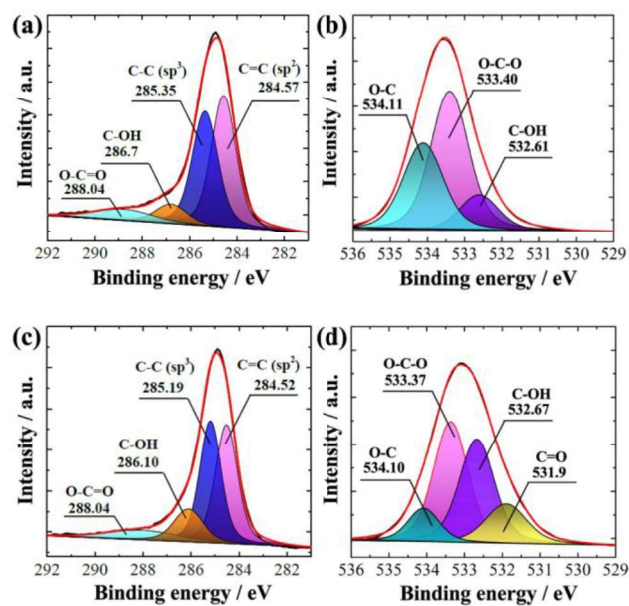


Figure S25: Raman spectra of pC<sub>70</sub>, FC, MCFC, and MCFC<sub>220</sub>.

## 27. X-ray photoelectron spectroscopy (XPS) studies



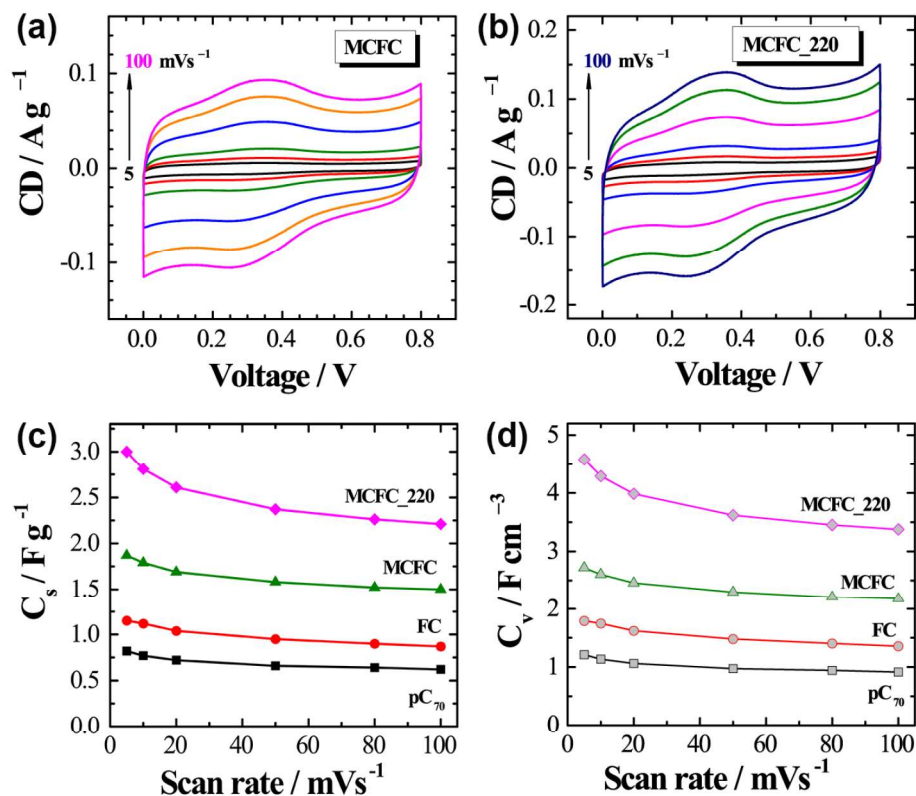
**Figure S26:** Survey XPS spectra of pC<sub>70</sub>, FC, MCFC, and MCFC<sub>220</sub>.



**Figure S27:** (a) C1s core level XPS spectra of FC with peak fit, (b) the corresponding O1s XPS core level spectrum with peak fit, (c) C1s core level XPS spectra with peak fit for MCFC, and (d) the corresponding O1s XPS core level spectrum with peak fit for MCFC.

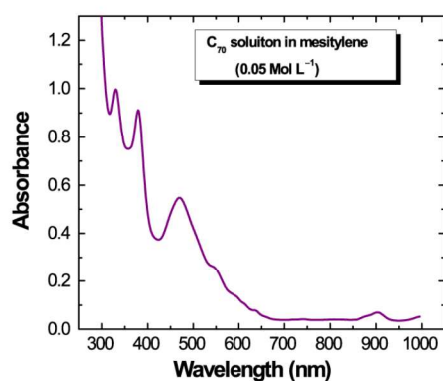


## 28. Cyclic voltammetry (CV)



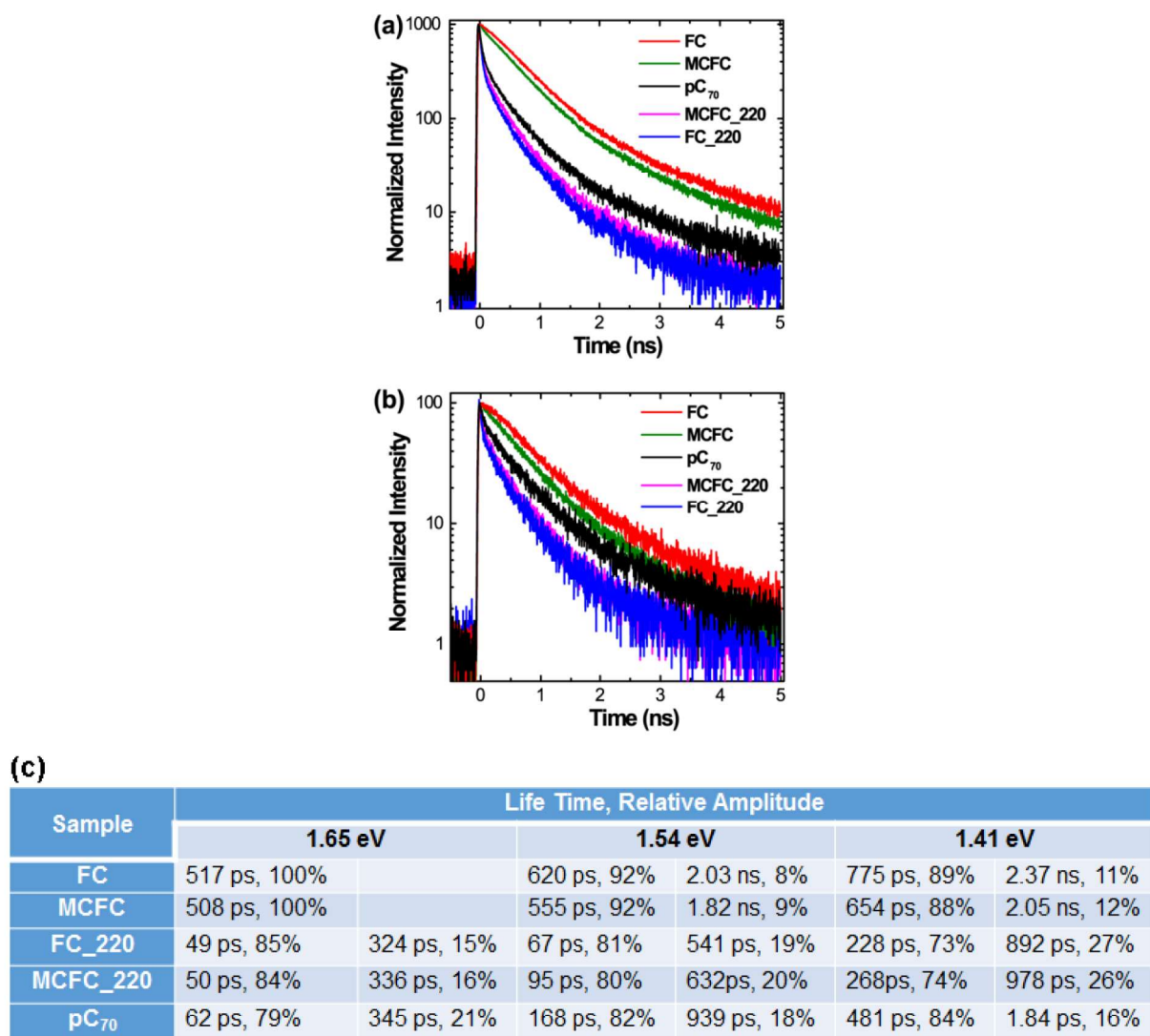
**Figure S28:** (a) CV curves of MCFC vs Ag/AgCl in 1 M H<sub>2</sub>SO<sub>4</sub> aqueous electrolyte at different scan rates (5, 10, 20, 50, 80, and 100 mVs<sup>-1</sup>), (b) corresponding CV curves of MCFC\_220, (c) calculated gravimetric specific capacitances (C<sub>s</sub>) of pC<sub>70</sub>, FC, MCFC, and MCFC\_220, and (d) corresponding volumetric specific capacitances (C<sub>v</sub>) pC<sub>70</sub>, FC, MCFC, and MCFC\_220.

## 29. UV-Vis spectrum of C<sub>70</sub> solution



**Figure S29:** UV-Vis spectrum of C<sub>70</sub> solution in mesitylene (0.05 mol L<sup>-1</sup>).

### 30. Additional TRPL data



**Figure S30:** (a) TRPL spectra measured at peaks around 1.54 eV for pC<sub>70</sub>, FC, FC<sub>220</sub>, MCFC, and MCFC<sub>220</sub>. (b) TRPL spectra measured at peaks around 1.41 eV for pC<sub>70</sub>, FC, FC<sub>220</sub>, MCFC, and MCFC<sub>220</sub>. (c) Summary of the time constants measured at 1.65, 1.54, and 1.41 eV, respectively.