Supplemental Information

Synthesis of Cu-Doped Mn₃O₄@Mn-Doped CuO Nanostructured Electrode Materials by a Solution Process for High-Performance Electrochemical Pseudocapacitors

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Measurements

The crystallographic phase of the compounds was studied by X-ray diffractometer (XRD, D8 Advance, Bruker, Germany) with a Cu K_{α} radiation ($\lambda = 1.5406$ Å). The morphologies were analyzed using a field-emission scanning electron microscope (FE-SEM, Hitachi S-4800, Japan) and a high-resolution transmission electron microscope (HR-TEM, Tecnai G2TM F20, Japan). Selected area diffraction pattern (SAED) and the lattice fringes of the samples were characterized by HR-TEM. Energy dispersive spectra (EDS) of elements were measured with an EDS analyzer (INCAx-sight7421, Oxford Instruments, UK) equipped with the FE-SEM. X-ray photoelectron spectra (XPS) were obtained with an XPS analyzer (XPS, Thermo Scientific[™] K-Alpha, Thermo Fisher Scientific, UK). Raman spectroscopic measurement (Horiba Scientific, Xplora Plus, France) was performed at room temperature with an excitation wavelength of 532 nm. Thermal properties were measured using a Scinco TGA-N 1000 analyzer in the temperature range between 30 to 800 °C in the presence of air. The optical absorption spectra were obtained using a UV-Visible spectrophotometer (Perkin Elmer, Lambda 35, USA). Fourier-transform infrared (FTIR) spectra were recorded on a Nicolet iS5 spectrophotometer (ASB1100426, ThermoFisher Scientific, Massachusetts, USA). The N₂ adsorption-desorption isotherm was obtained at -190 °C after evacuation at 155 °C for 10 hours using a surface area analyzer (Micromeritics, Tristar II 3020, USA).



Figure S1: N₂ adsorption-desorption isotherm of MO, CO, and CMO@MCO (reaction time 10 hours).



Figure S2: EDS spectrum of CMO@MCO prepared with the reaction time of 10 hours.



Figure S3: XPS survey spectra of MO, CO, and CMO@MCO (reaction time 10 hours).



Figure S4: XRD pattern of CMO@MCO prepared with the reaction time of (a) 5 and (b)15

hours.

Calculation of specific capacitance (C_s):

From the CV plots, the C_s of the electrodes was calculated conferring to the following equation (SI).

$$C_{s} = \frac{\int_{V_{1}}^{V_{2}} i(V)dV}{2 \times v \times (V_{2} - V_{1}) \times m}$$
(SI)

where, m is the total mass of active materials (ca. 50 mg for MO, CO, and CMO@MCO) $\int_{V_2}^{V_1} i(V) dV$ is the total voltammetric charge, and V₂-V₁ is the width of the potential in V, ν is the scan rate in V/s.

From the CD plots, the C_s values of the electrodes was calculated based on the following equation (SII).

$$C_{s} = \frac{I \times \Delta t}{A \times (V_{2} - V_{1})}$$
(SII)

where, I, and Δt are the galvanostatic discharge current, and discharge time obtained from the discharge plots, respectively.

Calculation of energy density (E_d) and power density (P_d):

The E_d and P_d of the electrode were measured using the following equations SIII and SIV, respectively.

Energy density (E_D) =
$$0.5 \times C_s \times (V_2 - V_1)^2 \times 0.2778$$
 (SIII)

Power density
$$(P_D) = E_D \times 3600 / \Delta t$$
 (SIV)



Figure S5: CVs of CMO@MCO/CC electrodes in 1 M KCl_(aq.) electrolyte at a scan rate of 100 mV/s with the reaction time of (a) 5, (b) 10, and (c) 15 hours for the synthesis of CMO@MCO.



Figure S6: CVs of (a) MO/CC, (b) CO/CC, and (c) CMO@MCO/CC electrodes in 1 M KCl_(aq.) electrolyte at the scan rates of 5, 10, 20, 30, 40, 50, 60, 70, and 80 mV/s.



Figure S7: Crystal structures of MO along the (101), (112), and (211) planes with tunnel structures, which was drawn by using Mercury software (version 4.2.0). The unit of the distance shown in the structures is Å.



Figure S8: Crystal structures of CO along the (200), (110), and ($20\overline{2}$) planes with tunnel structures, which was drawn by using Mercury software (version 4.2.0). The unit of the distance shown in the structures is Å.



Figure S9: CD plots of (a) MO/CC, (b) CO/CC, and (c) CMO@MCO/CC electrodes in 1 M KCl_(aq.) electrolyte at varying applied current densities.