Supporting online Materials for

p-nitroaniline redox-active solid-state electrolyte for battery-like electrochemical capacitive energy storage combined with asymmetric supercapacitor based on metal oxides functionalized β-polytype porous silicon carbide electrodes

Myeongjin Kim^a, Jeeyoung Yoo^b and Jooheon Kim^{a,*}

^a School of Chemical Engineering & Materials Science, Chung-Ang University, 211 Heukseok-dong, Dongjak-gu, Seoul 156-756, Republic of Korea

^b Program in Nano Science and Technology, Graduate School of Convergence Science and Technology, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul, Republic of Korea E-mail: jyoo78@snu.ac.kr

*Corresponding author: Jooheon Kim, <u>Tel:+82-2-820-5763</u>; Fax:+82-2-812-3495; E-mail address: jooheonkim@cau.ac.kr (J. Kim)

1. Calculations and equations

The ionic conductivity of electrolyte can be calculated by the following equation:

$$\sigma = \frac{1}{R_b \times A}$$
 S(1)

Where σ is the ionic conductivity (S cm⁻¹), l is the distance between the two stainless steel sheets (cm), A is the contact area of the electrolyte film with stainless sheets during the experiment (cm²), R_b is the bulk resistance (Ω).

The specific capacitances (C_s) are calculated from the CV and galvanostatic charge/discharge curves using the following equation:

$$C_{s} = \frac{1}{v(V_{c} - V_{a})} \int_{V_{a}}^{V_{c}} I(V) dV$$
 S(2)

$$C_{s} = \frac{I \times t}{m \times \Delta V}$$
 S(3)

where C_s is the specific capacitance (F g⁻¹), v is the potential scan rate (mV s⁻¹), $V_c - V_a$ represents the sweep potential range (V), I(V) denotes the response current density (A g⁻¹), I is the discharge current, t is the discharge time, m is the mass of active material, and ΔV is the voltage drop upon discharging.

The frequency response capacitances were estimated using a series-RC circuit model:

$$C(f) = \frac{-1}{2\pi f M \times Z''(f)}$$
 S(4)

where C is the capacitance (F/g), f is the frequency, M is total mass of active materials in both electrodes (g), and Z''(f) is the imaginary part of the impedance.

The frequency-dependent imaginary (C''(f)) component of the capacitance were obtained from the impedance spectra according to the following equation:

$$C''(f) = \frac{Z'(f)}{2\pi f M \times |Z(f)|^2}$$
 S(5)

where C''(f) is the imaginary capacitance, f is the frequency, M is total mass of active materials in

both electrodes (g), Z'(f) is the real part of the impedance, respectively, and |Z(f)| is the absolute value of the impedance.

The time constant (τ_0), which is a characteristic parameter indicating the rate capability of an electrical system, can be estimated from the peak frequency of C''(f) using the following equation:

$$\tau_0 = \frac{1}{f}$$
 S(6)

where f is the frequency corresponding to the peak frequency of the imaginary capacitance.

The power density and energy density of symmetrical supercapacitor systems were calculated using the following equations:

$$E = \frac{1}{2}C(\Delta V)^2$$
 S(7)

$$P = \frac{E}{t}$$
 S(8)

where *P*, *C*, ΔV , *t*, and *E* represent the power density (W kg⁻¹), specific capacitance based on the mass of the electroactive material (F g⁻¹), cell voltage for charging and discharging (V), discharge time (s), and energy density (W h kg⁻¹), respectively.



Figure S1. (a) Low-magnification FE-TEM image of SiCF. (b). High-magnification FE-TEM image of SiCF.



Figure S2. (a) Deconvoluted XPS Mg 2p spectra of SiCF/MgCo₂O₄. (b) Deconvoluted XPS Co 2p spectra of SiCF/MgCo₂O₄.



Figure S3. (a) Nitrogen adsorption-desorption isotherm of SiCF. (b) Pore size distribution of SiCF.



Figure S4. (a) Deconvoluted XPS Fe 2p spectra of SiCF/Fe₃O₄. (b) Deconvoluted XPS Fe $2p_{3/2}$ spectra of SiCF/Fe₃O₄.



Figure S5. Comparative CV curves of SiCF/Fe₃O₄ and SiCF/MgCo₂O₄ electrodes performed in a three electrode cell at a scan rate of 5 mV s⁻¹.



Figure S6. Specific capacitances for SiCF/MgCo₂O₄//SiCF/Fe₃O₄ cell with KOH aqueous and PVA-KOH gel electrolyte at different scan rates.



Figure S7. (a) CV curves of KOH and KOH-PNA aqueous electrolyte with SiCF electrode performed in a three electrode cell at a scan rate of 5 mV s⁻¹. (b) Gavanostatic charge/discharge curves of KOH and KOH-PNA aqueous electrolyte with SiCF electrode performed in a three electrode cell at a current density of 0.5 A g⁻¹.



Figure S8. Ionic conductivity of PVA-KOH-PNA gel electrolyte containing different amounts of PNA (PVA = 1 g, KOH = 1 g).