Electronic Supplementary Information (ESI)

Ribbon-like activated carbon with a multi-structure for supercapacitors

Mok-Hwa Kim,^{a,b} Kwang-Bum Kim,^b Kisuk Kang,^c Joong Tark Han^d and Kwang Chul Roh^{*a}

^a Energy & Environmental Division, Korea Institute of Ceramic & Engineering & Technology,

Seoul 153-801, Republic of Korea. E-mail: rkc@kicet.re.kr

^b Department of Materials Science & Engineering, Yonsei University, Seoul 120-749, Republic of Korea

^c Department of Materials Science & Engineering, Seoul National University, Seoul 151-742, Republic of Korea

^d Nano Carbon Materials Research Group, Korea Electrotechnology Research Institute, Changwon 642-120, Republic of Korea

1. Electrochemical measurements

The specific capacitance C_m in farads per gram of electrode ($\mathbf{F} \cdot \mathbf{g}^{-1}$) for all samples was calculated from the slope of the discharge curve using

$$C_m = \frac{I\Delta t}{\Delta V m},\tag{1}$$

where *I* is the discharge current (A), Δt is the discharge time period for the potential change in seconds, ΔV is the voltage difference during the discharge (V), and *m* is the mass of a single electrode (calculated on the basis of the total weight of the electrode materials).

The volumetric specific capacitance C_V was evaluated from

$$C_m = \frac{I\Delta t}{\Delta V_W},\tag{2}$$

where w is the volume of a single electrode.

The volumetric specific capacitances C_s were calculated from the C-V curves using

$$C_{s} = \frac{\int I_{s} \Delta V_{s}}{v \, \Delta V_{s} w},\tag{3}$$

where I_s is the response current, ΔV_s is the potential window (V), and v is the potential scan rate (V·s⁻¹).

The energy and power densities were calculated using the following formulas:

$$E = \frac{C_s(\Delta V)^2}{2}, \text{ and}$$
(4)

$$P = \frac{E}{t},\tag{5}$$

where *E* is the specific energy density ($W \cdot h \cdot kg^{-1}$), *P* is the specific power density ($W \cdot kg^{-1}$), and *t* is the discharging time (s). All of the calculations were consistent with those in references 8, 15 of the main text.

2. Supporting figures

Figure S1 shows the wide-angle XRD pattern for the samples. All samples exhibit a broad peak at $2\theta = 20-25^{\circ}$, which corresponds to the (002) reflection. The (002) peak gradually shifts from 20.6° (A600) to 24.5° (A900) and becomes slightly sharper with a higher carbonization temperature. The average values for the interlayer distance are listed in the figure caption. The average interlayer distance decreases with an increase in the temperature.



Figure S1. Effect of different carbonization temperatures on the structure of samples: (a) XRD pattern of raw pitch carbonized at different temperatures and (b) activated carbon structure resolved by XRD indicating a decrease in the interlayer distance with increasing in carbonization temperature. The interlayer distances d_{002} (nm) are as follows: A600 = 0.43, RAC = 0.39, and A900 = 0.36.

Pore characteristics for the samples are analyzed using the BET method. Figure S2 shows the nitrogen adsorption isotherms for the samples. In the isotherms of all samples, a large amount of N_2 is adsorbed at a remarkably low relative pressure (<0.1 P/P_o). It is noted that this isotherm type is in good agreement with the microporous carbon material structure.

The pore characteristics of both activated carbons are summarized in Table S1. It is found that A900 has nearly no pores, and its specific surface area is merely $3.36 \text{ m}^2\text{g}^{-1}$. Therefore, we confirm that the formation of pores in activated carbons depends on the graphitic structure of the carbonized sample and the effect of chemical activation.



Figure S2. N_2 adsorption/desorption isotherms for samples prepared at different carbonization temperatures.

Table S1. Pore characterization and specific capacitances of samples compared with a commercial activated carbon (MSP20, Kansai Coke & Chemicals, Japan).

	Specific	Total pore	Pore ratio (%)		Specific
Sample	surface area	volume	micro	meso &	capacitance
	(m^2g^{-1})	(cm^3g^{-1})		macro	$(\mathbf{F} \cdot \mathbf{cc}^{-1})$
A600	2513	1.12	84.0	16.0	35.4
RAC	1177	0.55	79.7	20.3	61.4
A900	3.36	0.01	10.3	89.7	45.6
MSP20	2272	1.01	90.0	10.0	36.8

The morphology of sample was investigated using field emission scanning microscopy (FE-SEM, JSM-6700F, JEOL, Japan). FE-SEM images of RAC are shown in **Figure S3**. There are no notable differences in the shape between the RAC and the other samples.



Figure S3. FE-SEM images of carbon materials: (a) raw pitch, (b) GC carbonized at 750 °C, and (c) RAC with chemical activation at 900 °C.

HR-TEM analysis is performed in order to examine the correlation between the structure of the materials to the specific capacitance. In **Figures S4 (a,b)**, the sample prepared by carbonization at 600 °C reveals disordered graphene layers and the formation of porous activated carbon. In contrast, HR-TEM micrographs of RAC and A900 indicate the existence of an ordered graphitic structure. **Figures S4 (c,d)** show images for C900 and A900; however, completely crystalline graphite is not formed. A900 exhibits an orderly graphitic crystalline arrangement. Carbonization at 900 °C promotes the growth of a graphitic crystalline arrangement and inclines it toward arranging. It should be noted that micro-explosions could not disarrange the structure during the activation process. This indicates that increasing the carbonization temperature increases the order of the sample, and no drastic structural changes occur by chemical activation. The specific capacitance of each sample is shown in **Figure S5**. The capacitance of RAC is much higher than those of A600 and A900. This is because of the progression of shallow intercalation-deintercalation; it is surmised that the performance is improved because additional capacitance is realized in the EDL capacitance of the porous structures. Furthermore, the RAC is compared with commercially activated carbons and exhibits the highest capacitance among commercial activated carbons.



Figure S4. HR-TEM images of samples showing the evolution of the partial crystalline structure according to increasing carbonization temperature: (a) carbonization at 600 °C (C600); (b) C600 with chemical activation at 900 °C (A600), where the A600 image shows the amorphous structure of graphene layer; (c) carbonization at 900 °C (C900); and (d) C900 with chemical activation at 900 °C (A900), where the A900 image exhibits an orderly graphitic crystalline arrangement.



Figure S5. Specific capacitance results for RAC relative to A600, A900, and MSP20.

Figure S6 show the results of cyclic voltammetry tests for RAC in a three-electrode system. Oxidation and reduction peaks are broadly observed, as indicated in the figure. It is considered that the electrolyte ions can intercalate into the distorted graphene layer structure derived from the process of interlayer expansion and shrinkage.



Figure S6. CV results measured at sweep rate of 5 mV·s⁻¹ in TEABF₄/PC within the potential range: 0-2.7 V (vs. Li/Li⁺).



Figure S7. (a) XRD patterns of the GC and RAC, (b) comparison of the (100)/(101) diffraction peaks in GC and RAC, (c) comparison of the Raman spectra of GC and RAC, and (d) comparison of the 2D peak for GC and RAC.

Figure S8 shows the close relationship for the voltage plateau appearing in the first charge cycle for various graphitic carbon materials.



Figure S8. Plot of the voltage plateau appearing in the first charge cycle for various graphitic carbon materials.