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Retransformed graphitic activated carbon from an ionic liquid-derived nitrogen containing carbon

Mok-Hwa Kim,[a,b] Sol Yun,[d] Ho Seok Park,*[d] Joong Tark Han,[c] Kwang-Bum Kim,[b] and Kwang Chul Roh*[a]

[a] Energy & Environmental Division, Korea Institute of Ceramic Engineering & Technology, Seoul 153-801, Republic of Korea

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

[c] Nano Carbon Materials Research Group, Korea Electrotechnology Research Institute, Changwon 642-120, Republic of Korea

[d] School of Chemical Engineering, Sungkyunkwan University, Suwon 440–746, Republic of Korea

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*Corresponding author. Tel.: +82 2 3282 2463 Fax: +82 2 3282 2475. E-mail address: rkc@kicet.re.kr (K. C. Roh).

METHODS

Material preparation

The activated carbon was prepared via the carbonization and activation of an ionic liquid (IL), namely 1-ethyl-3methylimidazolium dicyanamide (EMIM-DCA, Sigma Aldrich). This entailed carbonizing the IL at 900 °C for 1 h in N_2 to produce what is hereafter denoted as IL-C, which was then mixed gravimetrically with potassium hydroxide (KOH) to a 1:4 ratio. This alkali-rich residual carbon was then neutralized with 0.1 M hydrochloric acid (HCl) solution, filtered, and washed with distilled water until the pH reached a value of 6–7. The sample was then dried at 150 °C for 12 h to obtain an activated material referred to as IL-AC.

Characterization

To analyze the pore structure, samples were first outgassed at 200 °C for 18 h under vacuum. A gas analyzer (Belsorp-Mini II, BEL Japan Inc.) was then used to obtained N_2 adsorption-desorption isotherms, from which the specific surface area was calculated using the Brunauer-Emmett-Teller (BET) method. The graphitic structure of the samples was analyzed using X-ray diffraction (XRD, Rigaku D/Max 2500/PC, Japan), low angle XRD (Rigaku D/MAX 2200V/PC, Japan), high-resolution transmission electron microscopy (HRTEM, JEM 2000EX, JEOL, Japan) and Cs-corrected scanning tunneling electron microscopy (STEM, JEM-ARM 200F, JEOL). Raman spectroscopy was also conducted using a LabRam HR (Horiba Jobin Yvon, France) with an Ar-ion laser at 514.5 nm. The resulting Raman spectra were fitted using a Gaussian curve to determine the peak intensities and shifts. The atomic concentration of critical elements was determined by X-ray photoelectron spectroscopy (XPS) at 1.1×10^{-7} Pa, using a PHI 5000 VersaProbe with Al Kα radiation (ULVAC-PHI, Inc., Japan).

CO_2 adsorption capacity testing

Prior to testing, all samples were degassed under vacuum at 200 °C for 18 h. Their CO_2 adsorption was then tested at 25 °C using a Belsorp-HP volumetric adsorption analyzer (BEL Japan, Inc.).

Electrochemical measurements

Rubber-type electrodes were made using polytetrafluoroethylene (PTFE, D-60, Daikin Industries, Japan) as a binder and Super-P black (MMM Carbon Co., Belgium) as a conducting agent, giving a final composition of 90 wt% active material (IL-AC), 5 wt% conducting material and 5 wt% binder. These electrodes were then punched to 12 mm in diameter and dried in a vacuum oven to give a mass loading per area of 5.3 mg cm^{-2}. For comparative purposes, electrodes were also prepared in the same manner using as the active material a commercial activated carbon known to deliver the highest capacitance of all commercial activated carbons (MSP20, Kansai Coke & Chemicals, Japan).

The electrochemical characteristics were examined using a stainless-steel two-electrode cell containing an electrolyte of 1.0 M tetraethylammonium tetra-fluoroborate ($TEABF_4$) in propylene carbonate. Stainless steel 2032-type coin cells with two symmetrical electrodes separated by a porous polymer were assembled inside an Ar-filled glovebox, and were subjected to galvanostatic charge-discharge tests using a battery tester (Maccor, Series 4000, USA). Cyclic voltammetry tests were performed from 0 to 2.7 V using a potentiostat (EC-Lab, France), with all electrochemical tests being conducted at room temperature. The specific capacitance of a single-electrode in the symmetric two-electrode cell was calculated from the galvanostatic charge-discharge curve according to the equation:

$$C_{single\text{-}electrode} = 4 \times I/(m\ dV/dt)$$

where $C_{single\text{-}electrode}$, I, m, and dV/dt represent the specific capacitance for a single-electrode, the constant applied current, the active material mass of both electrodes, and the slope obtained from discharge curve, respectively.

Table S1. Comparison of the pore characteristics of IL-C, IL-AC and a commercial activated carbon (MSP20, Kansai Coke & Chemicals, Japan).

Sample	S_{BET}[a]	V_T[b]
IL-C	4	0.02
IL-AC	2823	2.30
MSP20	2272	1.01

[a] Specific surface area ($m^2 \cdot g^{-1}$) [b] Total pore volume ($cm^3 \cdot g^{-1}$)

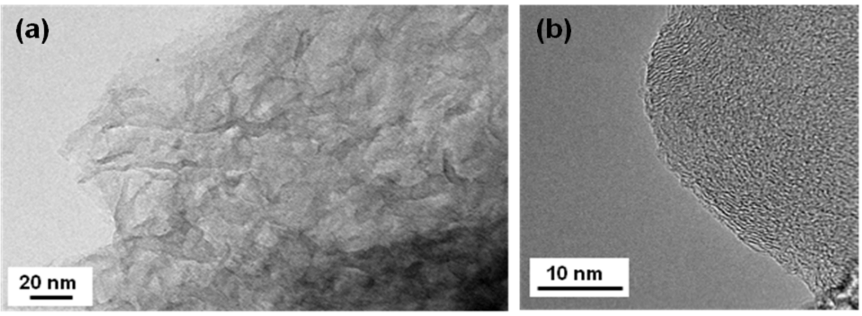

Fig. S1 (a) HRTEM images of IL-C showing its graphitic structure with limited extension. (b) STEM image of IL-C.

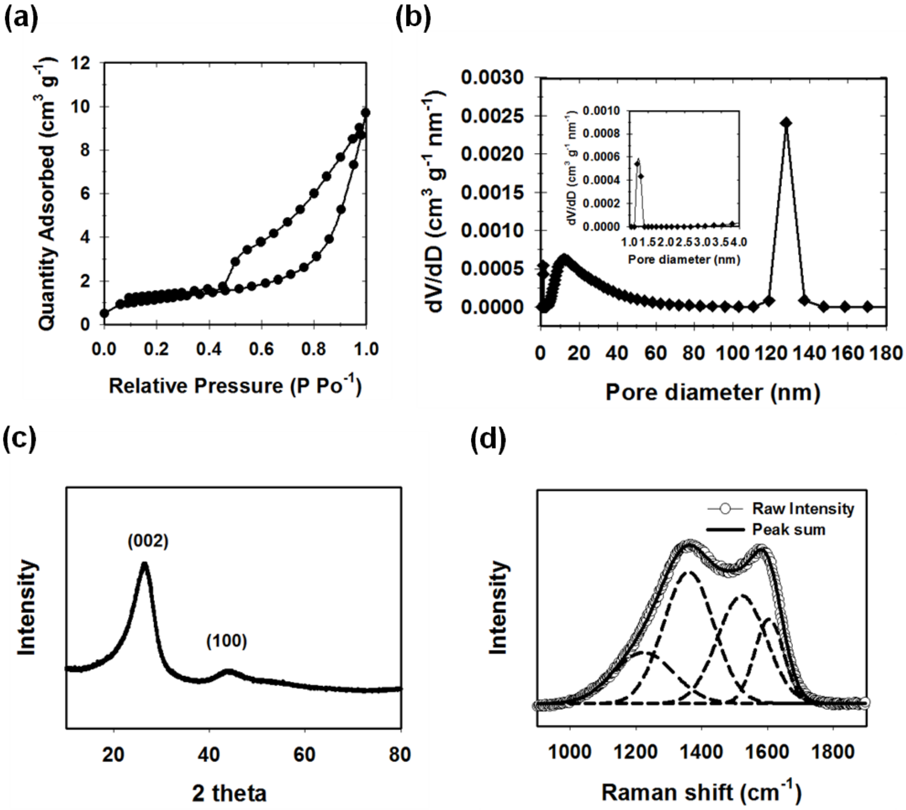

Fig. S2 (a) Nitrogen adsorption-desorption isotherms of IL-C. (b) Pore size distribution (PSD) curves of IL-C, as determined by an NLDFT model. (c) XRD pattern of IL-C showing the characteristic (002) and (100) planes or carbon. (c) Raman spectra of IL-C with deconvoluted peaks showing the four Gaussian sub-bands.

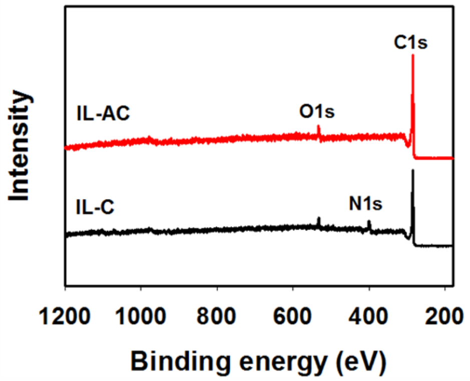

Fig. S3 XPS wide-spectra of IL-C and IL-AC.

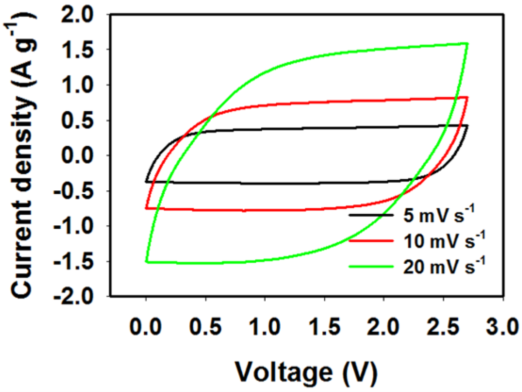

Fig. S4 Cyclic voltammetry curves of IL-AC at different scan rates.

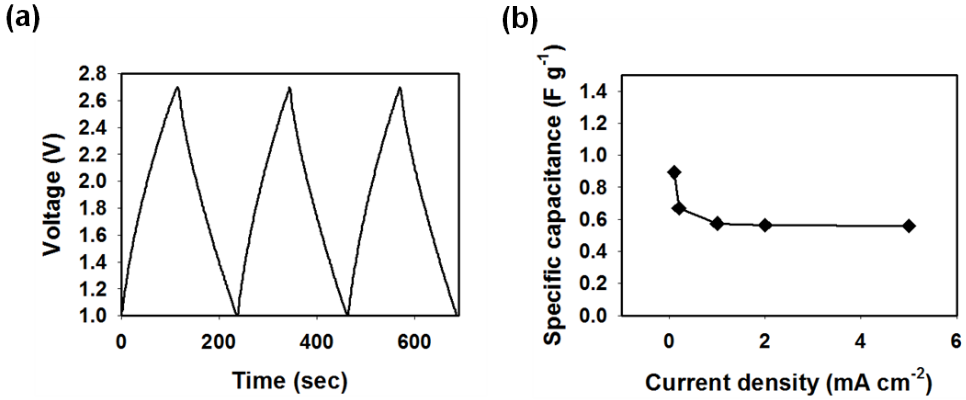

Fig. S5 Capacitive performance of IL-C: (a) galvanostatic charge/discharge profiles and (b) specific capacitance versus current density.