Electronic Supplementary Information

Novel Geopolymer Route to Porous Carbon; Effective CO$_2$ Adsorption Capacity

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Experimental Section

Materials: Kaolin (K7737) was purchased from Sigma-Aldrich. Water glass (SiO$_2$ 29%, NaOH 12.3%), HCl (35%), sucrose (98%), and H$_2$SO$_4$ (98%) were obtained from Daejung Chemicals. P123 (triblock copolymer EO$_{20}$PO$_{70}$EO$_{20}$, Pluronic 123) was purchased from BASF and HF (49%) from J.T. Baker. They were all used with no further purification.

Synthetic method: Ewha porous geopolymer-1 (EPG-1) was synthesized using P123 as the structure directing agent and kaolin as the aluminosilicate source. Kaolin was dehydroxylated at 750 °C to form metakaolin, and then successively dissolved in a solution of water glass with the pH of 13.5 at room temperature for 30 minutes. The resulting viscous aluminosilicate emulsion was mixed with an aqueous HCl solution of P123 with the pH ≤ 1.0, where the compositional molar ratio for Al:(Si+Al):P123:HCl:H$_2$O was 0.25:1.00:0.017:5.70:220. The mixture was then stirred for 12 hours at 40 °C, successively subjected to hydrothermal treatment at 100 °C for 24 hours, and cooled down to room temperature before filtering and drying to get the P123/EPG-1 composite, which was finally calcined in air at 550 °C to remove P123 to get EPG-1 designed. And then Ewha porous carbon-1 (EPC-1) was replicated from EPG-1 via the well-defined impregnation method using sucrose as the carbon precursor.[7] 1 g of EPG-1 was added to a solution in which 0.96 g of sucrose and 0.11 g of H$_2$SO$_4$ were dissolved in 5 g of water, and thus prepared suspension was stirred on a hot plate for 6 h at 60 °C. And the dough like phase was further kept in oven at 100 °C for 6 h, and at 160 °C for another 6 h. Finally the sample was ground in a mortar to obtain the powder form of sucrose impregnated EPG-1. To realize complete impregnation of sucrose in the pores of the EPG-1 template, we have done the same process twice as follows; 0.6 g of sucrose and 0.07 g of H$_2$SO$_4$ were dissolved in 5 g of water, which was again added to the sucrose impregnated EPG-1 powder previously

prepared. And the suspension was subjected to the stirring and thermal treatment as described above. It was then followed by pyrolyzation in a nitrogen flow at a heating rate of 5 °C min^{-1} up to 900 °C, and kept for 6 h to realize complete carbonization. After cooling to room temperature, the thus prepared EPG-1/EPC-1 composite was dispersed in 5 wt % hydrofluoric acid by stirring for 5 h to dissolve EPG-1 into solution. The undissolved porous carbon (EPC-1) was washed out several times with distilled water, filtered and finally dried at 100 °C.

Small angle x-ray scattering spectroscopy: The Rigaku SWXD (D/MAX 2500, 18 kW) Cu Kα radiation (λ = 1.5418 Å) was operated at 40 kV and 200 mA. In case of EPG-1, the diffraction peaks were characteristic of the hexagonal *p6mm* symmetry. The *d*-spacing was calculated to be 8.34 nm from (100) diffraction ($2\theta_{100}$ = 1.05 degree) on the basis of Bragg law (eq.1). The lattice constant *a* was calculated to be 9.62 nm based on the hexagonal type lattice (eq.2). Finally the wall thicknesses of EPG-1 was calculated to be 2.42 nm from the *a* value derived and D_{Pore} according to the structure (see Scheme S2).

$$n \, \lambda = 2d \, \sin\theta \ (1)$$

$$\frac{1}{d^2_{hkl}} = [\frac{4}{3} * (h^2 + k^2 + hk) + l^2 * \left(\frac{a}{c}\right)^2] * \left(\frac{1}{a^2}\right) \ (2)$$

Transmission electron microscopy: The TEM images were obtained using a JEOL JEM-2100F with an accelerating voltage of 200 kV that was used also for the energy dispersive X-ray spectroscopy (EDS) analysis for EPG-1.

Field-emission scanning electron microscope (FE-SEM): The FE-SEM images were obtained using a JEOL JSM-6700F with an accelerating voltage of 10 kV.

Low-voltage high-resolution scanning electron microscope (LVHR-SEM): All the images were taken on a JEOL JSM-7900F GBSH-S equipped with an upper electron detector (UED). The carbon stub was used as sample supporter. In order to induce efficient contact with samples, the carbon stub was polished with sandpaper (#1200) and successively filter paper, further cleaned by ultra-sonication in ethanol, and finally heated in vacuum to remove the alcohol. The samples dispersed in ethanol were dropped onto the stub, and then dried in vacuum oven for 10 minutes at 200 °C. Finally, it was cleaned using Ar ion cleaner equipped with a load lock chamber of the LVHR-SEM system at 300 V for 10 minutes. All the images were taken with a landing energy of 500 V after applying a specimen bias of -5 kV, and the gun voltage of 5.50 kV.

N$_2$ adsorption/desorption method: Nitrogen physisorption measurement was performed at 77 K on a BELSORP II mini instrument. BET method was used to calculate the surface area (P/P_0 = 0.05−0.20). And total pore volume was evaluated at P/P_0 = 0.99. The mesopore sizes were determined via BJH method. We also analyzed the porosity through nonlocal density functional theory (NLDFT)-based analysis using the WIBU-KEY software. All the samples were degassed for 5 hours at 200 °C before measurements.

X-ray fluorescence (XRF) spectroscopy: The chemical composition of kaolin was determined by XRF analysis using a Rigaku ZSX Primus IV.

CO$_2$ adsorption: The CO$_2$ adsorption was carried out using high-pressure Quantachrome Isorb HP1

instrument equipped with a water circulation system for the temperature control. The experiment was conducted at 273 K under pressures up to 30 atm.
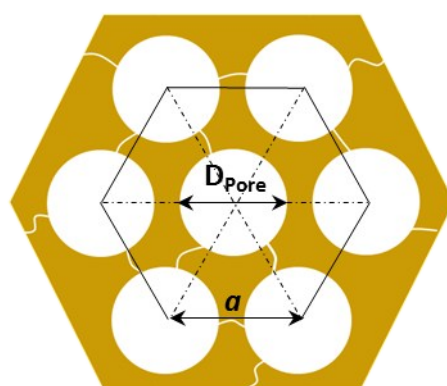
Table S1 Chemical composition of Kaolin.

Kaolin composition (wt. %)							
SiO_2	Al_2O_3	TiO_2	Fe_2O_3	P_2O_5	K_2O	CaO	LOI*
50.02	45.45	1.72	1.07	1.06	0.23	0.22	0.23

*LOI: Loss on ignition.
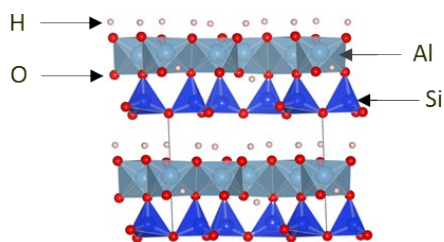
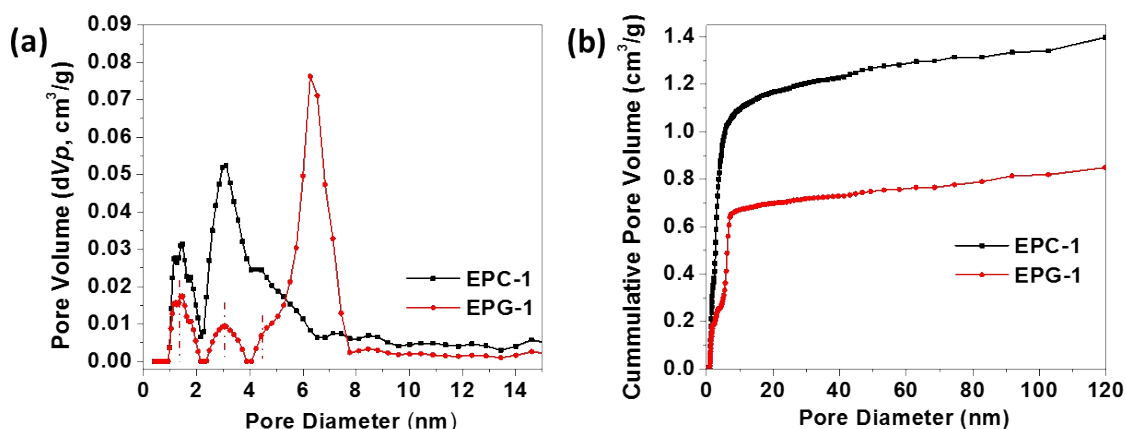
Scheme S1 Structure model of the 1:1 type aluminosilicate, kaolinite.

Wall thickness = a - D_{Pore}

Scheme S2 Structure model of Ewha porous geopolymer-1 (EPG-1).

Fig. S1 (a) Pore size distributions based on nonlocal density functional theory (NLDFT) based analysis and (b) cumulative pore volumes with respect to NLDFT method for Ewha porous geopolymer-1 (EPG-1) and Ewha porous carbon-1 (EPC-1).

Table S2 Textural porosities of Ewha porous geopolymer-1 (EPG-1) and Ewha porous carbon-1 (EPC-1).

	S_{BET}[1] (m^2/g)	D_{Pore}[2] (nm)	V_{Total}[3] (cm^3/g)	V_{Meso}[4] (cm^3/g)	V_{Micro}[5] (cm^3/g)	Mesoporosity[6] (%)	Microporosity [7] (%)
EPG-1	711	7.20	0.85	0.56	0.19	65	22
EPC-1	1551	2.70	1.41	0.92	0.34	65	24

[1]Specific surface area calculated from the Brunauer–Emmett–Teller equation.

[2]Main pore diameter derived from the Barrett, Joyner, and Halenda (BJH) plot.

[3]Total pore volume at the adsorption value of $P/P_0 = 0.99$.

[4]Cumulative volume in the mesopore range of 2–50 nm derived from the nonlocal density functional theory (NLDFT) method from Fig. S1 (b).

[5]Cumulative volume in the micropore range below 2 nm derived from the nonlocal density functional theory (NLDFT) method from Fig. S1 (b).

[6]Mesoporosity = $(V_{Meso} / V_{Total}) * 100 \%$

[7]Microporosity = $(V_{Micro} / V_{Total}) * 100 \%$

Table S3 CO_2 adsorptivity of Ewha porous carbon-1 (EPC-1) and other reported porous carbon materials from literature.

	CO_2 adsorption (mmol/g)	Temperature (k)	Pressure (atm)	Refs.
EPC-1	3.60	273	1	This work
	26.30	273	30	This work
CMK-3	1.55	303	1	[1]
	3.20	273	1	[2]
	10.45	298	30	[3]
	16.70	273	30	[4]
	17.50	275	30	[5]
	24.40	273	30	[4]
Activated carbon	3.66	273	30	[6]
Multiwalled carbon nanotubes	5.63	273	30	[6]
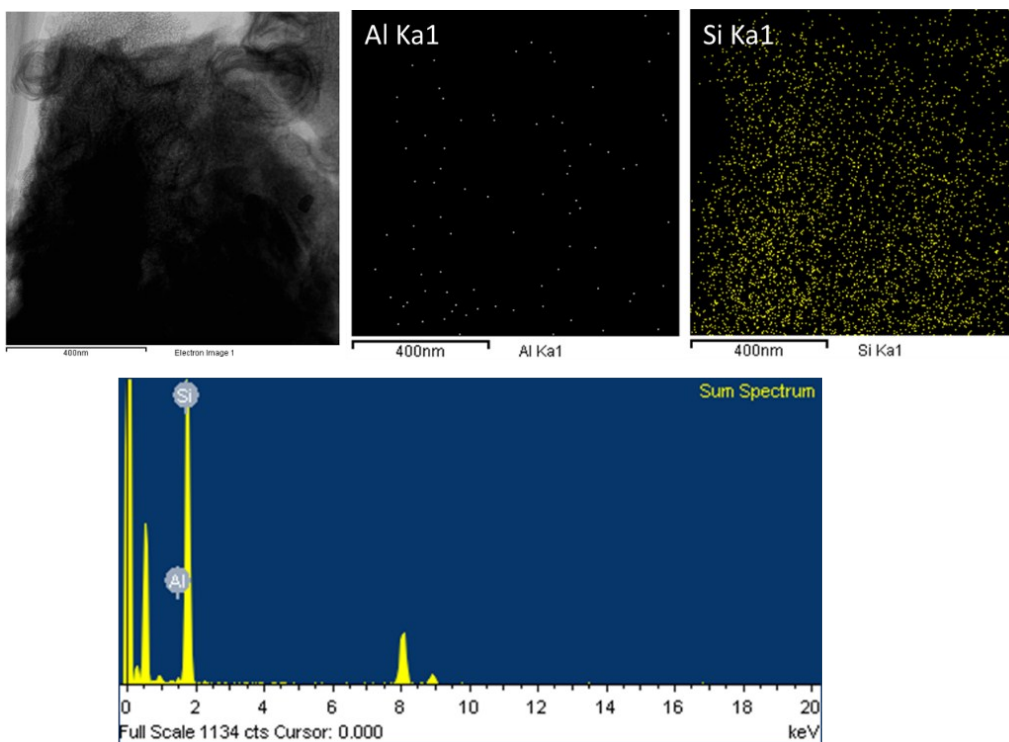
Fig. S2 EDS analysis results for Ewha porous geopolymer-1 (EPG-1).

Table S4 EDS analysis results for Ewha porous geopolymer-1 (EPG-1).

Element	Weight %	Atomic %
Al K	0.99	1.03
Si K	99.01	98.97
Totals	100.00	100.00

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