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Short communication

Electrochemical performance of hybrid supercapacitor fabricated using multi-structured activated carbon

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ABSTRACT

We used multi-structured activated carbon electrodes to fabricate hybrid supercapacitors (HSCs) showing excellent electrochemical performance. As the active material, the activated carbon coupled with $Li_4Ti_5O_{12}$ showed a partially graphitized as well as a porous structure, and thus provided two different capacitive mechanisms: electric double layer capacitance and shallow intercalation. HSCs produced with this unique structure show excellent specific capacitances of 77 F g^{-1} and 62 F cm^{-3}. Partially graphitic activated carbon has great potential for HSC applications.

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1. Introduction

With the recent full-scale commercialization of power generation and distribution systems such as renewable-energy-based systems, smart grids, and hybrid electric vehicles, there has been an increasing interest in energy-storage devices showing superior electrochemical performance that can support such systems [1]. Supercapacitors are highly efficient devices that exhibit high power densities (1–5 kW kg^{-1}) and nearly unlimited cycle life provided by a charge-storage mechanism involving non-faradaic double-layer capacitance. However, the use of supercapacitors has been limited because of their low energy densities (<10 Wh kg^{-1}) [2].

To achieve high-energy-density supercapacitors, studies have focused on hybrid supercapacitors (HSCs) consisting of asymmetric electrodes that combine the advantages of faradaic and non-faradaic charge storage mechanisms of lithium ion batteries (LIBs) and electric double layer capacitors [3–6]. Previous studies have mainly focused on the performances of the LIB materials to improve the energy densities of HSCs, which are restricted by the low capacitance limits of carbonaceous materials.

Previously, we reported that a supercapacitor fabricated using partially graphitic activated carbon (PG-AC) exhibited an electric double-layer and shallow intercalation capacitances [7]. It showed a volumetric capacitance of 67% which is higher than commercial activated carbon. However, the material showed a lower rate capability than that of existing activated carbon owing to the additional mechanism of shallow intercalation [8].

The rate performance of HSCs depends on the electrode having a relatively low rate capability between asymmetrical electrodes. In the case of an HSC consisting of an LIB material and PG-AC, it will be determined by the rate capability of the LIB material with faradaic lithium-intercalation reaction because the rate capability of PG-AC mainly exhibits the non-faradaic capacitive reaction and is far superior to that of LIB materials. Thus, the use of PG-AC will not affect the rate performance of HSC and will improve the specific capacitance through the enhanced capacitance of the carbon electrode.

In this study, we therefore designed a new HSC with a high specific capacitance. The positive electrode material, PG-AC, was synthesized by the carbonization and alkali activation of pitch. The PG-AC electrode was coupled with spray-dried microspherical porous $Li_4Ti_5O_{12}$ as the negative electrode material because it showed good structural stability owing to its negligible volumetric change, high electrode density, and porous structure that facilitates electrolyte penetration. Under capacitance balancing between asymmetric electrodes, the PG-AC/

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$Li_4Ti_5O_{12}$ exhibited excellent specific capacitances of 77 F g^{-1} and 62 F cm^{-3}.

2. Material and methods

The partially graphitic activated carbon (PG-AC) was prepared using a previously reported method [7]. Isotropic pitch (Anshan Chemical Co., China) was carbonized at 750 °C for 1 h in an argon atmosphere. The carbonized sample was mixed with KOH, for activation, in a ratio of 1:4 (wt.%) and then heated at 900 °C for 1 h in an argon atmosphere. The resulting PG-AC was neutralized with diluted HCl, washed with distilled water, and dried at 100 °C for 24 h.

$Li_4Ti_5O_{12}$ was prepared by spray drying. Li_2CO_3 and anatase TiO_2 (60 nm) were used. The Li/Ti molar ratio was 4:5, and a dispersant (BYK-190) was mixed in distilled water for 12 h. A two-fluid nozzle atomizer was used to atomize the mixed slurry at 200 °C and 3.5 kg cm^{-2}. The spray-dried powder was then heated at 750 °C for 6 h in air.

The microstructures of carbonized sample and PG-AC were investigated using high-resolution transmission electron microscopy (HR-TEM, JEM-2000EX, JEOL). The morphology of the $Li_4Ti_5O_{12}$ was investigated using field-emission scanning electron microscopy (FE-SEM, JSM-6700F, JEOL). The crystalline structures were analyzed using X-ray diffraction (XRD, Rigaku-D/Max-2500/PC), and the specific surface area and pore distributions were measured using the Brunauer–Emmett–Teller method (BELSORP-max).

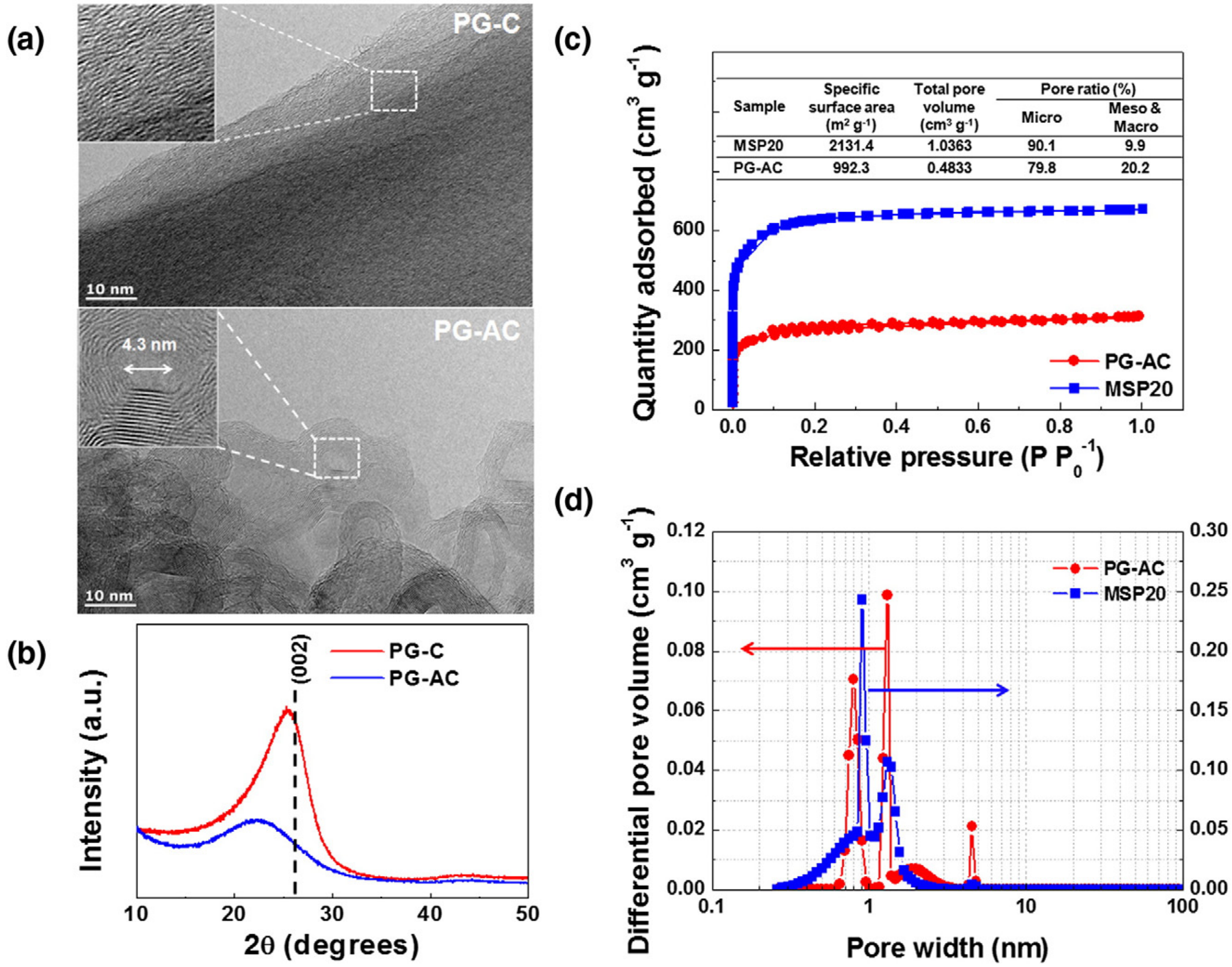
Rubber-type electrodes were fabricated using 90 wt.% active material, 5 wt.% conductive carbon, and 5 wt.% polytetrafluoroethylene (PTFE) binder to compare commercial activated carbon (MSP20, Kansai Chemical Co.) and PG-AC. The thickness and diameter of each electrode were controlled and punched to 120 μm and 12 mm, respectively.

The negative electrode was produced by coating an Al foil with a slurry containing prepared $Li_4Ti_5O_{12}$, conductive carbon, and polyvinylidene fluoride (PVDF) binder mixed in a weight ratio of 86:8:6. The $Li_4Ti_5O_{12}$ electrode was 25 μm thick.

Electrochemical tests were performed using a three-electrode cell (ECC-REF); the separator and electrolyte were Celgard®2400 and a 1 M solution of $LiPF_6$ dissolved in ethylene carbonate/ethyl methyl carbonate mixed in 1:1 vol.% ratio, respectively. Lithium metal was used as the counter and reference electrodes for the half-cell and three-electrode cell tests, respectively. The SUS discs of various thicknesses (1–10 mm) were used to control the positions of the separator and the reference electrode between positive and negative electrodes in ECC-REF. The half-cell tests of the positive electrodes were performed in the range of 3–4.45 V vs. Li^+/Li, and those of the negative electrodes were performed in the range of1–3 V vs. Li^+/Li. The HSCs were galvanostatically charged/discharged in the ranges of 0.2–6 mA cm^{-2} and 1.5–2.8 V.

3. Results and discussion

As shown in Fig. 1(a), the sample carbonized (PG-C) at 750 °C had a crystal structure consisting of partially parallel and randomly oriented



Sample	Specific surface area (m^2 g^{-1})	Total pore volume (cm^3 g^{-1})	Pore ratio (%)	
			Micro	Meso & Macro
MSP20	2131.4	1.0363	90.1	9.9
PG-AC	992.3	0.4833	79.8	20.2

Fig. 1. (a) HR-TEM images and (b) XRD patterns for PG-C and PG-AC; (c) nitrogen adsorption–desorption curves, and (d) pore size distributions for MSP20 and PG-AC.

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