Supporting Information

High-capacity, high-power organic electrode via supercritical CO_2 impregnation into activated carbon micropores

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Experimental

Materials and Chemicals

Activated carbon AC (Maxsorb® MSC-30) was purchased from Kansai Coke and Chemicals Co. Ltd. Chloranil (CHL) was purchased from Tokyo Chemical Industry Co. Ltd. Polytetrafluoroethylene (PTFE: 6-J) was purchased from Chemours-Mitsui Fluoroproducts Co., Ltd.

Impregnation of CHL into AC using $scCO_2$

The critical temperature and pressure of CO_2 were 31.1 °C and 7.38 MPa, respectively. CHL impregnation into AC was performed using supercritical CO_2 ($scCO_2$) in the apparatus shown in Figure 1. CHL (1 g) was placed in a SUS316 tube container (V:5.71 ml) with 0.1 g of AC. The AC was placed in a container composed of copper plates to prevent spillage. The container with CHL and AC was placed in a vat filled with aluminum balls. The vat was placed on a hot plate that can be heated at different temperatures (45–155 °C). Liquid carbon dioxide was introduced using a carbon dioxide transport pump (PU-2080-CO2 Plus, Jasco)that can be set to different pressures (10–25 MPa), and allowed to stand for 10 min. After stabilization at the desired pressure, valve 6 was closed, and the container was allowed to stand for 24 h.

Liquid impregnation of quinones into AC was used as a control. Chloranil (300 mg) was dissolved in acetone (100 ml) and sonicated for 10 min. AC (700 mg) was dispersed in the solution, followed by further sonication for 1 h. The solution was stirred at 60 °C so that acetone evaporated, and chloranil was impregnated into the pores of the AC.

Electrochemical measurement setup

AC impregnated with CHL was mixed with PTFE binder in a weight ratio of 9:1, and 5 mg of the mixture (mass loading: 21 mg/cm^2) was pressed to form pellets (Radius of the pellet ϕ: 5.5 mm)An aqueous solution of H_2SO_4 (0.5 M) bubbled with nitrogen for 2 h was employed as the electrolyte. The electrodes immersed in the electrolyte were placed under a vacuum for 30 min to remove bubbles.

The half-cell system consisted of CHL-impregnated AC as the working electrode, AC electrode (activated carbon, carbon black, and PTFE in a weight ratio of 8:1:1) more than ten times heavier than the working electrode as the counter electrode, 70 ml of aqueous H_2SO_4 solution (0.5 M) as the electrolyte, and an Ag/AgCl electrode as the reference electrode. Measurements were initiated using a potentiostat/galvanostat (Solartron Analytical) under ambient conditions. The capacity, cycle performance, and rate performance were evaluated using galvanostatic charge and discharge tests. When testing the half-cell performance, current density calculations were based on the weight of the entire electrode. The electrochemical impedance spectroscopy (EIS) was carried out in the frequency range from 10 Hz to 200 kHz with an amplitude of 10 mV.

Materials characterization

The specific surface area (SSA) was measured using an N_2 adsorption/desorption analyzer (Bellsorp Mini II), and the SSA was calculated from the adsorption points for relative pressures of 0.1–0.25 Pa using the Brunauer–Emmett–Teller (BET) theory. The pore distribution was measured using a pore size analyzer (QUADRASORB evo, Quantachrome Instruments). The pore size distribution was calculated using the nonlocalized density functional theory (NLDFT). Thermogravimetric measurements were performed using a TG/DTA 6200 (EXSTAR 6000 series, Seiko Instruments Inc., Japan) instrument. The temperature was increased from 30 to 110 °C (10 °C/min) under a N_2 flow of 200 ml/min. The temperature was maintained for 20 min, increased again to 400 °C (10 °C/min), and then kept constant for 40 min.

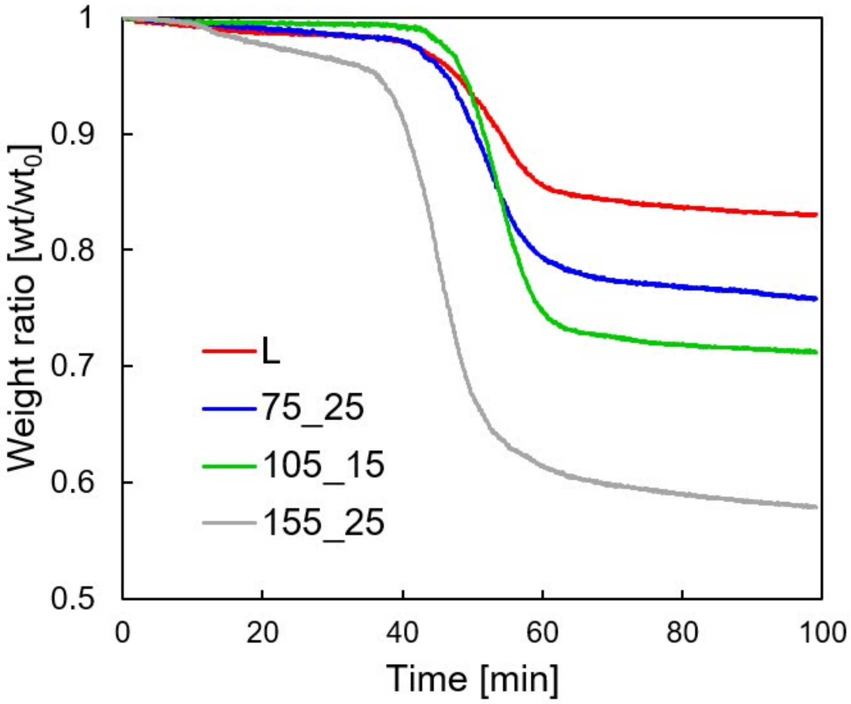

Figure S1. TG measurement results (under air atmosphere)

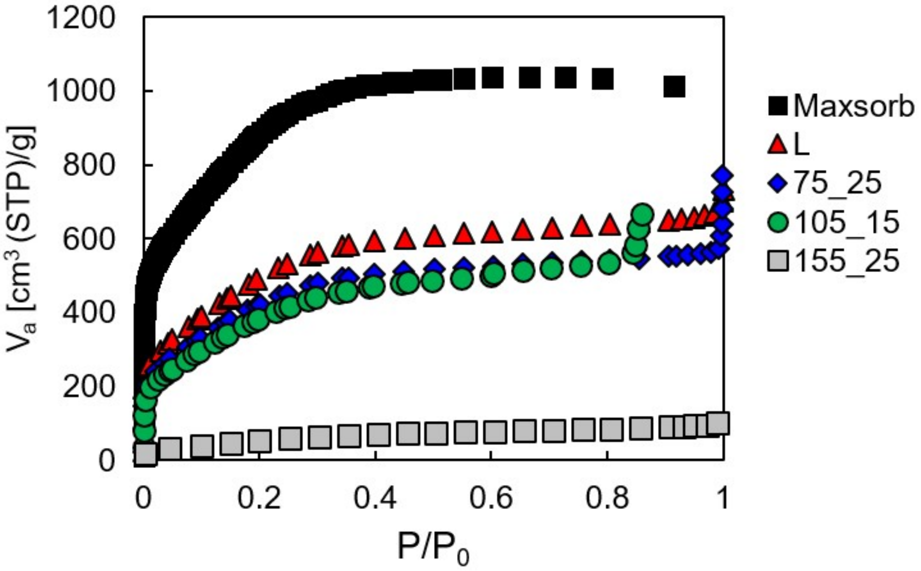

Figure S2. Adsorption isotherms for nitrogen adsorption measurements under characteristic samples.

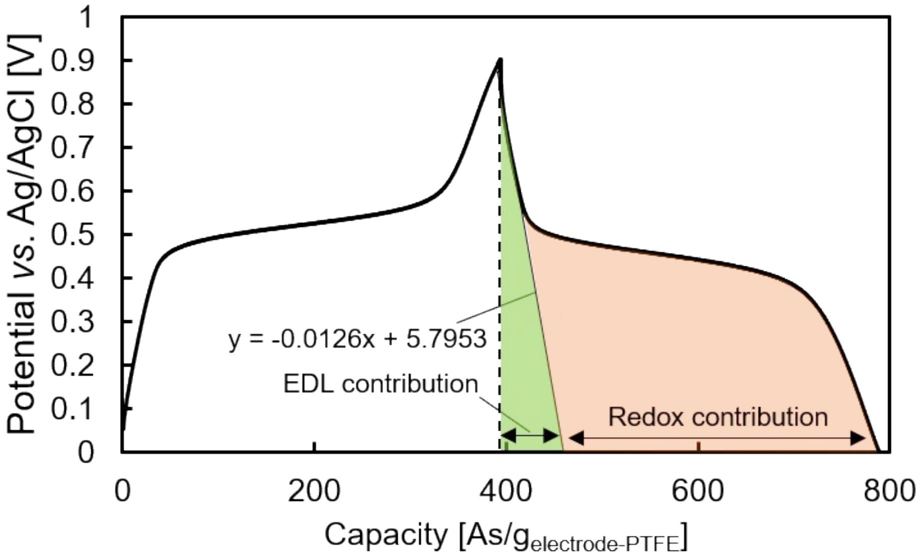

Figure S3. EDL and the redox contributions to the specific capacity of the CHL/AC electrode.

We describe the calculation method used to determine the redox capacity. The redox capacity of the CHL/AC electrode is calculated by subtracting the EDL capacity from the total discharge capacity, as reported previously. As an example, we calculated the redox capacity of the CHL/AC electrode prepared using $scCO_2$ impregnation at 105 °C and 15 MPa.

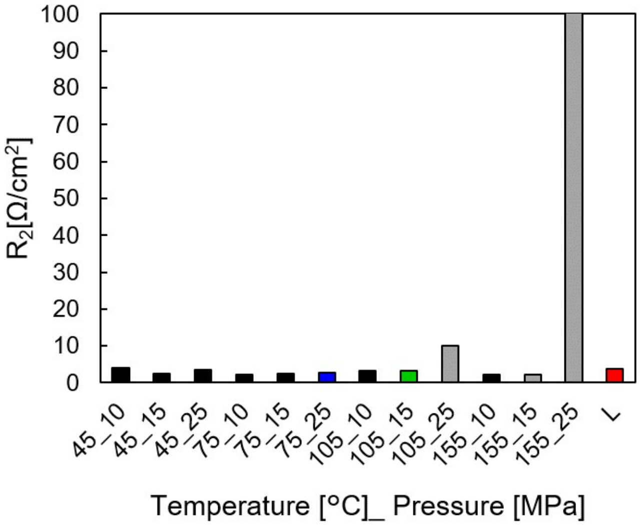

Figure S4. The results of EIS in CHL-impregnated AC electrodes prepared under different temperature-pressure conditions at 0.1 A/$g_{electrode-PTFE}$.