Secondary Activation of Commercial Activated Carbon and its Application in Electric Double Layer Capacitor

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Abstract: The cheap commercial activated carbon (AC) was improved through the secondary activation under steam in the presence of $FeCl_2$ catalyst in the temperature range of 800-950℃ and its application in electric double layer capacitors (EDLCs) with organic electrolyte was studied. The re-activation of AC results in the increases in both specific capacitance and high rate capability of EDLCs. For AC treated under optimized conditions, its discharge specific capacitance increases up to 55.65 F/g, an increase of about 33% as compared to the original AC, and the high rate capability was increased significantly. The good performances of EDLC with improved AC were correlated to the increasing mesoporous ratio.

Keywords: Activated carbon, electric double layer capacitor, improvement, high rate capability.

Recently, there has been an increasing interest in the development of EDLCs using highly porous carbons as the electrode materials due to their possible technological applications as energy storage components possessing high power density capability. The main advantages of such devices are their possible high rate capability and long cycle life as compared to rechargeable batteries, but their energy density is relatively lower than that of rechargeable batteries. The specific surface area and pore size distribution of AC have been considered to be two key factors to determine high energy and power density of EDLCs[1,2]. In particular, high mesoporous ratio of AC is highly beneficial to obtaining both high energy and power density of EDLC in organic electrolytes[2]. Recently, some researchers[3,4] have focused on the preparation and modification of carbon materials in order to obtain desirable specific energy and power density of EDLCs. In this paper, we focused on the improvement of cheap commercial AC and its application in EDLC with high rate capability.

The commercial activated carbon (Kureha Chemical Co. Japan) with a specific surface area of *ca.*1050 m^2/g was used as the starting material. Improvement on commercial AC was performed through secondary activation under water steam in the presence of $FeCl_2$ catalyst in the temperature range of 800-950°C. To evaluate the electrochemical performances of EDLCs with ACs, sheet-type electrode pellets

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(geometric area: 2 cm^2) and screw-type cells were employed. A pair of polarizable electrodes separated by one glass fiber was placed face to face. The screw-type cells were assembled in an argon-filled glove box. Within the polarizable electrodes, the weight ratio of AC : conductor : polyvinglidene fluoride (PVdF) is 8:1:1, where PVdF served as the binder. Each electrode contains about 12 mg of AC. The electrolyte used was 1.0 mol/L Et_4NBF_4 in propylene carbonate. Cyclic voltammetric measurements were performed between –3.0 and 3.0V at different scan rates using a Solartron 1286 Electrochemical Interface in two electrode system. Unless stated otherwise, the screw-type cells were charged and discharged at the current density of 0.4mA/cm^2 with the cut-off voltage of 0 to 3.0V. All the electrochemical measurements were performed at about 30°C.

Figure 1 shows the galvanostatic charge-discharge curves with two kinds of AC samples at 50th cycle. By comparison of the curves in **Figure 1**, it can be seen that the charge-discharge curves exhibit the typical capacitive behavior and the capacity of improved AC is higher than that of the original AC, indicating that the capacity of AC was increased after the improvement. For AC treated at 900°C for 1 hr, its discharge specific capacitance at 50th cycle is about 55.65 F/g, which shows an increase of more than 33% as compared to the original AC. Furthermore, the specific capacitive values of all AC samples improved in the temperature range of 800-950°C are higher than that of untreated AC.

Figure 1 The 50th charge-discharge curves for polarizable electrodes fabricated with (a) AC improved at 900°C for 1 hr and (b) original AC

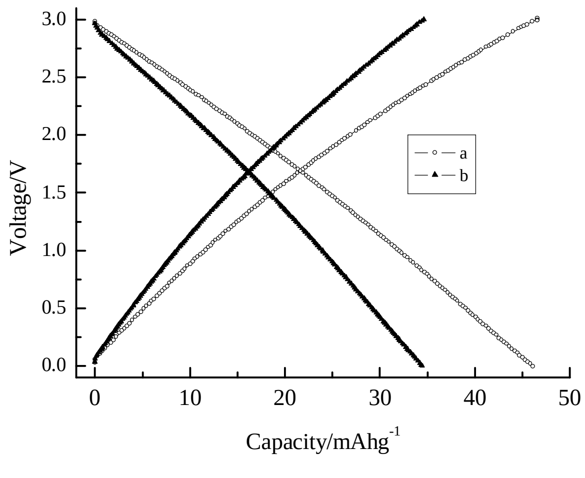
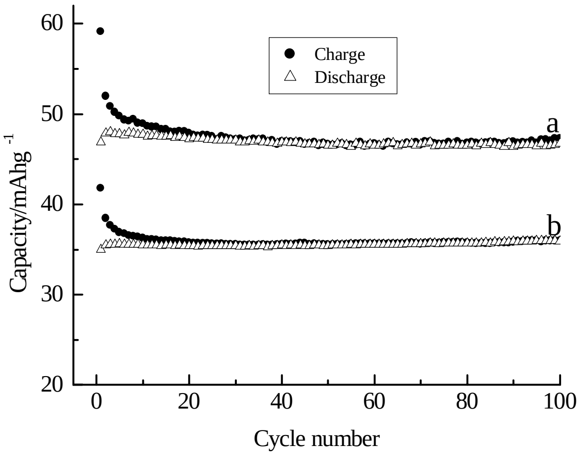


Figure 2 shows the cycling performance of EDLCs made with two AC samples. As seen in the figure, the capacity at any cycle for improved AC is higher than that for untreated AC, and the coulombic efficiency is clear to 100% for the two kinds of AC materials. Considering the specific capacitance and high rate capability of EDLCs as well as the yield during the secondary activation process, the suitable improvement

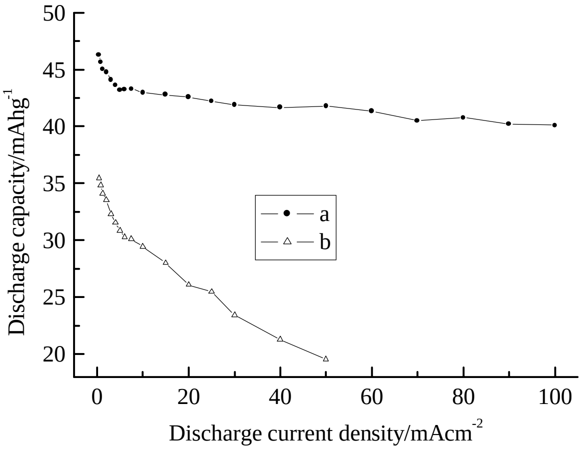
conditions for AC should be heating treatment at about 900°C for 1 hr.

Figure 2 The cycling performances of EDLCs made with (a) AC improved at 900°C and (b) original commercial AC



It is well known that the main interest of AC-based EDLCs with suitable specific energy is their capability to possibly deliver high specific peak power. **Figure 3** shows the dependence of discharge capacity for two kinds of AC samples on the discharge current density. As shown in the figure, the discharge capacity for EDLC with the original AC continuously decreases with the increase of discharge current density. But, for EDLC with improved AC, its discharge capacity slightly decreases with the increase of discharge current density when the current density is less than *ca.*10 mA/cm^2. When the current density is more than 10 mA/cm^2, its discharge capacity almost keeps constant. These results clearly proved that EDLC with improved AC as active materials can be cycled at very high current densities and the improved AC seems to be a suitable material for the fabrication of EDLC with high power density. Furthermore, from the cyclic voltammetric results, EDLC with improved AC exhibits good rectangular-shape voltammograms (not shown in this paper) at different scan rates. But for EDLC with the original AC, good rectangular-shape voltammogram was observed only at low scan rate. Meanwhile, the specific capacitive values of EDLC with improved AC decreases from 77.04 F/g to 65.34 F/g with the increase of scan rate from 10 mV/s to 200 mV/s. But for EDLC with original AC, its specific capacitive value decreases from 42.21 F/g to 18.61 F/g with the increase of scan rate from 10 mV/s to 200 mV/s. These results definitely proved that high rate capability of EDLCs was significantly increased after AC was improved. For the cylindrical EDLC with the volume of 13 cm^3 with improved AC as active material, its discharge power density is about 1.60 kW/kg when the EDLC released 85% energy. The high rate capability of such EDLCs should be ascribed to the increasing mesoporous ratio of AC after improvement, which will be reported in detail in another paper.

Figure 3 The dependence of specific discharge capacity of polarizable electrodes with (a) AC improved at 900°C for 1 hr and (b) original AC on the dischargecurrent density



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