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Electrochemical properties of mesoporous carbon aerogel electrodes for electric double layer capacitors

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Electric double layer capacitors (EDLC) have been extensively investigated in the recent years as an intermediate power source between conventional capacitors and rechargeable batteries. Electric double layer capacitors (EDLCs) with high power density are among the most important development in recent years in the field of energy storage and conversion, which could be used in applications such as memory back-up devices or electric vehicles. EDLCs store energy in the electric double layer by charge accumulation at the interface between the electrode and the electrolyte. In recent years, porous carbon materials such as activated carbon powder [1–7] and fiber [8], are the most frequently used electrode material for EDLCs. Because of poor conductivity of these materials, researchers show great interests in finding new electrode materials with high conductivity to improve energy and power density of EDLCs. With the characteristics of high conductivity and better pore size, carbon aerogels and carbon nanotubes were considered as the most potential electrode materials for EDLCs. However, some disadvantages of carbon nanotubes, such as high cost, low specific surface area and specific capacitance, had limited its applications for EDLCs. Carbon aerogels with the advantages of both aerogels and carbon materials have larger specific surface area, larger pore size and high conductivity, in 1994, so LLNL [9] had predicted that carbon aerogels would be widely used in electrochemical fields. A previous study [10] had shown that conductivity of the carbon aerogels is relatively stable between a wide temperature ranges (from 50 K to 300 K), carbon aerogels are suitable to be used as electrodes materials for EDLCs, in which electrolytes can be chosen according to working temperature. In this paper, carbon aerogels were prepared through sol–gel method, using linear phenolic and furfural as raw materials. The relationship between structures and electrochemical properties of carbon aerogels was discussed.

The preparation method of the carbon aerogels was described as follows. A propylol solution of phenolic (linear soft point was 75 °C), furfural and catalyst (0.005 mmol/100 gPR) led to polycondensation and the formation of gel. The gel invested in this paper were dried in supercritical petroleum ether for 1 h, and transformed into carbon aerogels by pyrolysis at 600–1000 °C for 3 h.

Carbon aerogels (98 wt.%), and polytetrafluoroethylene (PTFE) (2 wt.%) were mixed in order to obtain a powder, and then the powder was pressed to the Nickel collector electrode. The electrode had a surface of 100 mm^2 and thickness of 0.4 mm. A polypropylene separator separated the two carbon electrodes. The electrolyte was a 30 wt.% KOH aqueous solution.

The dc capacitances of carbon aerogels were measured using a self-designed coin-like test cell. The measurements were carried out at 25 °C using Arbin Instruments BT-4$^+$ (America). The dc capacitance C’ in Farad was calculated from the following equation (1) [5]: $C' = (i \times \Delta t)/\Delta v$, the specific capacitance C in farad/gram was calculated from the following equation $C = 2(I \times t)/(\Delta V \times m)$, where I is the constant discharge current in ampere, t is the time for discharge in second, ΔV is the potential change of the capacitor caused by discharge in volt, and m is the single electrode mass.

The Transmission Electron Microscope (TEM) photographs of prepared carbon aerogels were shown in Fig. 1. Carbon aerogels consisted of a network of particles arranged in an interconnected chain-like structure. The diameter of particles was about several nanometers.

In order to research the relationship between structures and electrochemical properties of the samples, four different kinds of carbon aerogels (C598, C602, C729, C855) were prepared, whose structure parameters and electrochemical properties were listed in Fig. 2, Fig. 3 and Table I. Fig. 1 shows N_2 adsorption-desorption isotherms of four kinds of carbon aerogels. All four isotherms show a distinct type IV according to the IUPAC classification, which meant that the dominant pores in these samples were mesopores and macropores. It was considered that the increase of the adsorbed volume at the high relative pressure was due to the effect of pore saturation on the mesopores. The pore volume distributions of the mesopores for the four samples calculated by the adsorption isotherms using

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TABLE I Parameter of pore structure and electrochemical properties of carbon aerogels

CA	B.E.T. (sq. m/g)	Total pore volume (cm^3/g)	D (nm)	Specific capacitance (F/g)	Double-layer capacitance (F/m^2)
C598	598	1.43	10.23	193.6	0.32
C602	602	1.26	9.04	164.6	0.27
C729	729	1.58	8.67	202.2	0.28
C855	855	1.50	10.0	196.4	0.23

Average pore diameter: $D = 4\ V_{total}/S_{BET}$.

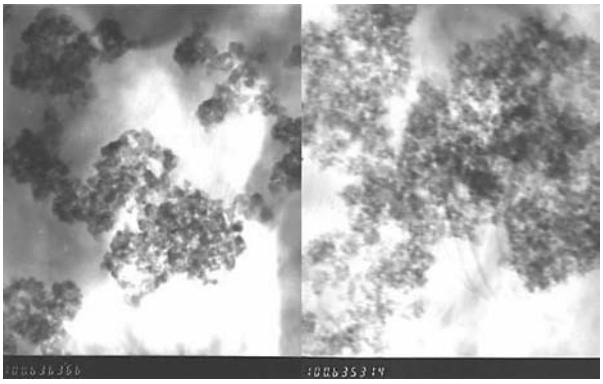

Figure 1 TEM photograghs of carbon aerogels.

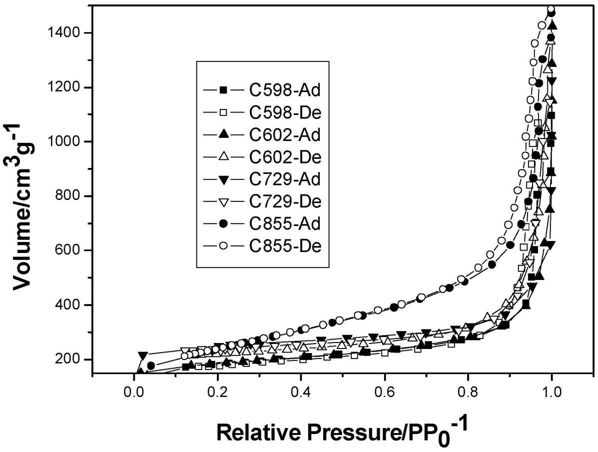

Figure 2 Nitrogen adsorption isotherms of activated carbons (Ad: adsorption; De: desorption).

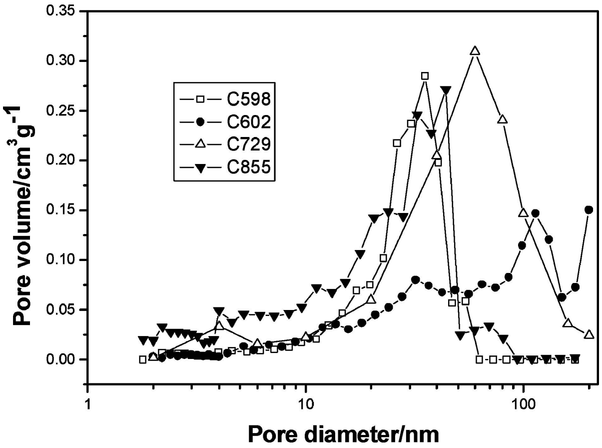

Figure 3 Pore volume distribution of activated carbons.

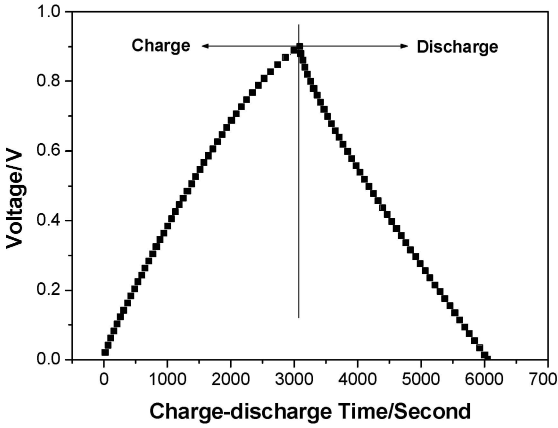

Figure 4 Discharge curves of the carbon aerogel electrodes with different current.

the Barret–Joyner–Halenda (BJH) method were shown in Fig. 3.

The data in Table I showed that C598 and C602 samples had almost the same specific surface but the significant difference in specific capacitance. Compared C598 with C855, the specific surface of C855 was larger than that of C598, but both had the similar specific capacitance. Although C729 had not largest specific surface in four samples, it had highest specific capacitor. These data indicated that there was not a direct relationship between the specific surface and the specific capacitance of carbon aerogels. A previous study [11] had shown that higher surface area of activated carbon means the larger specific capacitance, but the values of discharge capacitance of carbon aerogels did not increased with the increase in the BET surface area. Fig. 3 showed C598 and C855 had similar pore volume distribution, while two samples had similar specific capacitance. It could be seen that the C602 with the lowest pore volume had a low capacitance value. This must be due to the carbon aerogels merely provided the low surface area to form effective double layer. Compared with C602, C598 with relatively larger pore volume had higher specific capacitance, which showed that the larger pore volume led to higher capacitance while those surface areas were the similar. It could be seen that C729 had a largest pore volume and C602 had a smallest pore volume between 10 and 100 nm, so the specific capacitance of C729 was as high as 202.2 F/g, but the specific capacitance of C602 was only 164.6 F/g. It can also be concluded that the capacitance of carbon aerogels is determined by two factors, namely, surface area and total pore volume. Taking these two factors into consideration, larger pore volume gives the highest capacitance, especially between 10 and 100 nm was important to store energy for EDLC. It is well known that

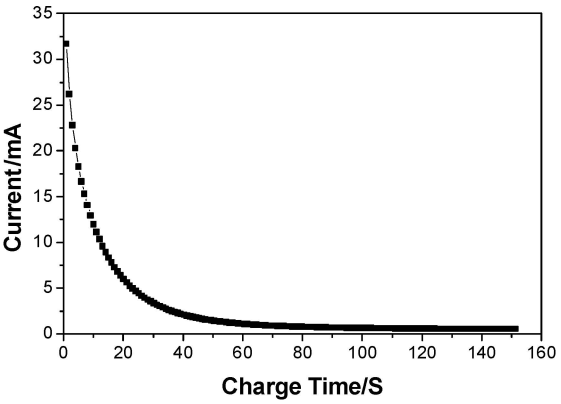

Figure 5 Relationship of charge current and time of carbon aerogel electrode under constant 0.9 V.

the double layer capacitance of pure graphite is around 0.2 F/m^2. Double-layer capacitances of four samples were more than that of pure graphite and decreased with the increase in the BET surface area. It was indicated that surface utilizing ratio decreased with the increase in the BET surface area. Compared with previous work, the mesoporous carbon aerogel samples possess higher capacitance, which is expected to be due to the unique mesoporous structure that provides OH$^-$ accessible pores and a very high surface area for charge storage.

Figure 4 depicted the charge and discharge curves of sample C855 at 0.5 mA currents. As shown in Fig. 4, voltages decreased linearly with increasing discharge time, moreover, the efficiency of charge–discharge kept 99%, which indicated that carbon aerogel electrodes have excellent charge-discharge properties. In Fig. 5 charge current and time of carbon aerogel electrode under constant 0.9 V represented, in 20 s of charge time, current rapidly decreased from 35 to 0.2 mA which had already approached to leak current. The characteristic of rapid charge–discharge ability under high current was proved. These electrochemical properties were very important to EDLCs.

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