Effect of Pore Packing Defects in 2-D Ordered Mesoporous Carbons on Ionic Transport

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Ordered mesoporous materials show great importance in energy, environmental, and chemical engineering. The diffusion of guest species in mesoporous networks plays an important role in these applications, especially for energy storage, such as supercapacitors based on ordered mesoporous carbons (OMCs). The ion diffusion behavior in two different 2-D hexagonal OMCs was investigated by using cyclic voltametry and electrochemical impedance spectroscopy. In addition, transmission electron microscopy, small-angle X-ray diffraction, and nitrogen cryosorption methods were used to study the pore structure variations of these two OMCs. It was found that, for the OMC with defective pore channels (termed as pore packing defects), the gravimetric capacitance was greatly decayed when the voltage scan rate was increased. The experimental results suggest that, for the ion diffusion in 2-D hexagonal OMCs with similar mesopore size distribution, the pore packing defect is a dominant dynamic factor.

1. Introduction

Ordered mesoporous materials have shown great potential in many important applications, such as chemical catalysis, environmental separation, and energy storage.1–3 All these applications involve the transportation of guest species (gas/liquid) in the mesoporous networks. Therefore, it is extremely vital to understand the effect of ordered mesopores on the dynamic processes of these guest species with respect to design of new ordered mesoporous structures.

Electrical double layer capacitors (EDLCs), in which the electrical energy storage/release depends on ion transport in the framework composed of nanometer-sized pores (mainly mesopores), have been considered as a promising energy storage device in many fields.2–5,8–12 Understanding the influence of different mesoporous materials (ordered or disordered arrangement) on ion diffusion behavior is fundamental to enhancing EDLC’s performance. However, in the past, the diverse and uncontrollable intrinsic microporous structure of physically or chemically activated carbons has made it impossible to precisely investigate the relations between mesoporous textures and ion diffusion behavior. Recently, Ryoo and co-workers have synthesized ordered mesoporous carbons (OMCs).6,7 These OMCs, with mesopore size tailorable and regularly interconnected mesoporous channels, are considered to be very promising materials for investigating the impact of ordered mesopores on ion diffusion behavior, which determines the energy density of EDLCs under large current operations.

A few groups have studied the impact of ordered mesopores on ion diffusion behavior by evaluating the performance of EDLCs based on OMCs.8–12 Their experiments show that 2-D hexagonal OMCs exhibit better EDL capacitive performance than 3-D cubic OMCs, which is believed to be attributed to the favorable ion diffusion in larger mesopores existing in 2-D hexagonal OMCs.8,9,12 Nevertheless, it is clear that only one-dimension diffusion is available in 2-D hexagonal OMCs. It can be assumed that if the single diffusion direction is blocked, no other diffusion routes will be available. Consequently, it is quite important to understand to what extent the blockage of the single diffusion direction will influence the ion diffusion ability of 2-D hexagonal OMCs.

A simple method was developed in this paper to control the blockage conditions in 2-D hexagonal OMCs. The capacitance-decaying trends of two different 2-D OMCs were investigated by cyclic voltammogram (CV) at different voltage sweep rates. Moreover, electrochemical impedance spectroscopy (EIS), which is a powerful method for obtaining information on the dynamic ion diffusion behavior,13–16 has been utilized to evaluate the influence of blockage conditions on the ion diffusion behavior in 2-D hexagonal OMCs. Combined with mesopore structure characterizations, the present results clearly demonstrate that defective 2-D ordered mesoporous channels dramatically reduce the ion diffusivity in mesopores.

2. Experimental Section

2.1. Synthesis and Characterization of OMCs. The 2-D hexagonal OMCs materials were obtained by a templating procedure for which ordered mesoporous silica SBA-15 was chosen as a template. The SBA-15 templates were prepared according to the method described by Jun et al.7 However, the final calcination process, which is used to remove the surfactant embedded in SBA-15, was altered to adjust the remaining surfactant amount. For Condition 1, the silica template was calcined in an oxygen-poor atmosphere and dark brown powder was obtained; for Condition 2, the silica template was calcined in an oxygen-rich atmosphere yielding pure white powder. The oxygen-poor atmosphere was realized simply by air-proofing the volume-limited oven tube, because the available oxygen...
amount in the volume-limited oven tube was exhausted during calcination. The oxygen-rich atmosphere was realized by controlling the inner pressure of the oven tube to be about double that of the atmospheric pressure under a flowing air stream. The effect of oxygen-poor atmosphere was to remove the surfactant in primary mesopores of the silica template, while keeping the surfactant in secondary mesopores. In the case of an oxygen-rich atmosphere, the surfactant in both primary and secondary mesopores was removed.

OMCs were hence synthesized by first templating the pretreated SBA-15 with sucrose as carbon precursor and then carbonizing by a prescribed heating procedure in argon atmosphere. The resultant silica/carbon composites were washed with NaOH dissolved in a deionized water/ethanol (volume ratio 1:1) (0.1 M) solution at 373 K for 3 h to remove the silica template. The remaining carbon was then filtered and rinsed until the pH was 7. Thereafter, the wet powder was dried under vacuum at 373 K for 12 h. The resultant carbon powders were denoted as CS-1 and CS-2 to differentiate the respective calcination conditions of the corresponding SBA-15 templates.

The OMCs were characterized by TEM (JEOL 2010, 200 kV). The OMCs samples were mixed in absolute ethanol and ultrasonically dispersed for several seconds. Then, the dispersed homogeneous solution was dropped onto a microgrid and dried for TEM observations. The small-angle X-ray diffraction (SAXRD) patterns were collected with use of an RINT2200 (Cu Ka, λ = 1.5406 Å) at a step scan rate of 0.02° from 1.5° to 10°. Nitrogen cryosorption was conducted on a Micrometrics ASAP2010M; before measurements, all the OMC samples were evacuated at 473 K until the manifold pressure was lower than 2 Pa. The external surface specific surface area (SSA) is the difference between BET specific surface area (BET-SSA) and micropore surface SSA. The BET-SSA was calculated based on the BET method. The micropore surface SSA was calculated based on the t-plot method. All these calculations were accomplished with the software provided along with the ASAP2010M instrument.

2.2. Electrochemical Measurements. The electrochemical measurements were carried out in a three-electrode system. Nickel foam covered with a composite of activated carbon powders and poly(tetrafluoroethylene) (PTFE) was used as counter electrode (CE). Hg/HgO was chosen as reference electrode (RE) in an alkaline electrolyte of 6 M KOH. The preparation of the working electrode (WE) for the three-electrode system was as follows. The active materials, CS-1 and CS-2, 5.5 mg each, were mixed with 1 mg of PTFE in absolute ethanol and spread onto nickel foam with a size of 10 mm × 10 mm, respectively. Then, the nickel foam covered with 5.5 mg of active materials was used as the WE. The electrolyte was 6 M KOH aqueous solution. The cyclic voltammogram and electrochemical impedance spectroscopy were collected on Solartron 1287/1260 electrochemical systems. The potential range for CV was −1 to 0 V vs Hg/HgO. The bias potential applied to the electrode during EIS was 0 V vs Hg/HgO, the frequency range was 10 mHz to 100 kHz, and the AC signal amplitude was 10 mV.

3. Results and Discussion

3.1. Pore Structure Characterization of OMCs. The TEM images of CS-1 and CS-2 perpendicular to their pore channels are illustrated in Figure 1a–d. It can be seen that the pore channels of CS-1 were far from parallel and straight compared to those of CS-2. During the TEM observations, it was found that most of the CS-1 samples possessed similar defective pore structure. In fact, sections 2 and 3 (2 and 3 in Figure 1a) indeed possess more defective pore structure than section 1 (1 in Figure 1a) because of the absence of distinct localized regular pore structure. However, considering the easy identification of defective pore structure, we chose to show the HRTEM of section 1 (1 in Figure 1a) to provide a distinct comparison between the regular pore structure and the defective pore structure, as observed in Figure 1b. Compared with the straight parallel mesopore channels in Figure 1d, the mesopores in Figure 1b comprise a large number of randomly distributed defective sites. The irregular defective sites in the mesopore channels mainly include (i) collapse of the primary mesopores and (ii) blockage of the primary mesopores.

It is accepted that the ordered hexagonal mesoporous framework composed of primary carbon rods in 2-D OMCs is supported by many smaller secondary carbon nanorods, and the absence of these short nanorods would cause an irregularly collapsed primary mesopore texture, as in the case of CS-1, which results from the unstable packing conditions of mesopore channels. The detailed formation mechanism of the irregularly collapsed mesopore channels of CS-1 can be explained as follows. As mentioned above, the SBA-15 template used for the preparation of CS-1 was calcined in an oxygen-rich atmosphere and showed a dark brown color after calcination, indicating incomplete removal of the surfactant, and the content of the carbon residue from surfactant was determined to be 6 wt % by TG in air up to 1273 K, shown in Figure IS (see the Supporting Information). It is important to mention that both SBA-15 templates possessed similar primary mesopore sizes, which probably resulted from complete removal of surfactant existing in the primary mesopores. Hence, the brown-looking carbon residue from surfactant must exist in the smaller secondary mesopores inside the silica walls. Consequently, during the preparation of CS-1, a large number of smaller inner-wall mesopores could not be impregnated by sucrose molecules due to the blockage by the carbon residue from surfactant. That condition would have led to a shortage of smaller secondary carbon nanorods to support the ordered mesoporous structure of resultant OMCs, and hence resulted in the irregular collapsing defects observed in the TEM images. Furthermore, the carbon...
order compared with that of the regular mesoporous channels of CS-2 and (2) CS-1 has a larger mesopore size than that of CS-2.

3.2. Electrochemical Performance of OMCs. The influence of ordered mesoporous structure on ion transport behavior can be characterized by the CV method based on the evaluation of capacitive performance of EDLC based on OMCs. Generally, the desired capacitive performance requires a rectangular shaped voltammogram. Besides, the capacitive behavior can also be studied by changing the voltage sweep rates to estimate the applicability for quick charge—discharge operations.

The cyclic voltammograms of CS-1 and CS-2 are presented in Figure 4, panels a and b, respectively, for different voltage scan rates. The voltammograms of these two carbons maintained the desired rectangular shape at voltage scan rates of 5 mV/s, where the charging and discharging curves were parallel with the voltage axis. When the voltage scan rate was increased to 50 mV/s, a quasirectangular shape can still remain for CS-2, indicating excellent capacitive behavior even at high current intensity. However, the voltammogram of CS-1 exhibited a triangular shape at 50 mV/s, indicating the absence of capacitive behavior at high current density. This means that CS-1 would be less suitable for quick charge—discharge operations than CS-2. The absolute gravimetric capacitance and specific surface capacitance (capacitance per unit surface area) are listed in Tables 1 and 2, respectively. From Table 3, it can be seen that CS-1 has higher specific surface capacitance than CS-2. This probably comes from the superior active surface of CS-1, as suggested by DSC results shown in Figure 2S (see the Supporting Information). As the voltage scan rates increased, the difference of gravimetric capacitance and specific surface capacitance between CS-1 and CS-2 decreased. The same trend was also observed from the results in a two-electrode system, as shown in Tables 1S and 2S (see the Supporting Information). This probably originates from the absence of capacitive behavior of CS-1 at high current density. To evaluate the capacitive behavior of CS-1 and CS-2, the ratio of retained gravimetric capacitance vs increased voltage scan rates is plotted in Figure 5. CS-2 maintains 82% of its capacitance at a high voltage scan rate of 50 mV/s, which is more than the ratio of 74% for CS-1. These unexpected results show a better capacitive behavior for CS-2 despite its smaller mesopore size than that of CS-1. Generally, the faster the penetration of electrolyte ions into electrochemically active porous surface, the better the capacitive behavior at high voltage scan rate. Comparing CS-1 with CS-2, the inferior capacitive behavior of CS-1 is unexpected because of its relatively larger mesopore size. However, as mentioned previously, the pore channel packing defects (PPDs) in CS-1 have higher specific surface capacitance than CS-2. This probably comes from the superior active surface of CS-1, as suggested by DSC results shown in Figure 2S (see the Supporting Information). As the voltage scan rates increased, the difference of gravimetric capacitance and specific surface capacitance between CS-1 and CS-2 decreased. The same trend was also observed from the results in a two-electrode system, as shown in Tables 1S and 2S (see the Supporting Information). This probably originates from the absence of capacitive behavior of CS-1 at high current density. To evaluate the capacitive behavior of CS-1 and CS-2, the ratio of retained gravimetric capacitance vs increased voltage scan rates is plotted in Figure 5. CS-2 maintains 82% of its capacitance at a high voltage scan rate of 50 mV/s, which is more than the ratio of 74% for CS-1. These unexpected results show a better capacitive behavior for CS-2 despite its smaller mesopore size than that of CS-1. Generally, the faster the penetration of electrolyte ions into electrochemically active porous surface, the better the capacitive behavior at high voltage scan rate. Comparing CS-1 with CS-2, the inferior capacitive behavior of CS-1 is unexpected because of its relatively larger mesopore size. However, as mentioned previously, the pore channel packing defects (PPDs) in CS-1 are much different from those in CS-2. To account for the larger mesopore size and inferior capacitive behavior for CS-1, it is believed that the different PPDs for CS-1 and CS-2 must have played a dominant role in determining the capacitive behavior at high voltage scan rates.

3.3. Electrochemical Impedance Spectroscopy of OMCs. Although the CV method can be utilized to estimate the ion transport behavior in ordered mesoporous structure by varying
the voltage sweep rates, it is still unable to precisely describe the actual electrochemical diffusion process. Hence, it is quite important to further investigate the influence of ordered mesoporous structure on ion diffusion based on EIS, which has been considered as a powerful method to obtain dynamic ion diffusion information.\textsuperscript{13}

The formation of EDL capacitance under an alternative electric field for a mesoporous/microporous electrode should involve three processes: (a) a high-frequency region where mass transfer is inhibited, so charge aggregation at the surface of carbon powder electrode in contact with the bulk electrolyte would be dominant; (b) a medium-frequency region where the dominant process would be ion diffusion in mesoporous channels which contributes the most to the development of capacitive behavior; and (c) a low-frequency region where inhomogeneous diffusion in the less-accessible sites (like micropores) may govern the impedance.\textsuperscript{13} Thus, the inferior dynamic capacitive performance of CS-1 at high voltage scan rates ought to be related to the ion diffusion ability in mesoporous channels which is identified as the medium-frequency region in both Nyquist and Bode plots.

The complex plane impedance plots (Nyquist plots) for CS-1 and CS-2 are given in Figure 6a. The knee frequencies are illustrated in the inset of Figure 6a, and the values for CS-1 and CS-2 are 39.8 and 63.1 Hz, respectively. Generally, the knee frequency is considered to be the critical frequency where EDLC begins to exhibit capacitive behavior. In the impedance plots, when the frequency is less than the knee frequency, a straight line, nearly vertical to the realistic impedance axis (\(Z\)'), was observed, characteristic of an admirable capacitive behavior. Deviation from the vertical line is attributable to inner-mesopore diffusion resistance for electrolyte ions, which is strongly dependent on the detailed mesoporous structure of the different samples. Therefore, it is possible to investigate the dynamic process of ion diffusion in ordered mesopores based on EIS.

In the phase angle plot, the approach to pure capacitive behavior at low frequency is usually identified with phase angle approaching to the negative 90 degree.\textsuperscript{14} Accordingly, the value of the phase angle can be used to evaluate the effectiveness of ion diffusion in mesopores at the medium-frequency region. That is, the smaller the phase angle, the better the capacitive performance and, hence, the faster the ions diffuse. The frequency dependent behaviors of the phase angle, \(\Phi\), are illustrated in Figure 6b (Bode plots). When the frequency is lower than 1.6 Hz, where the impedance behavior of both CS-1 and CS-2 would be ion-diffusion controlled, the phase angle of CS-2 is always smaller than that of CS-1. This result indicates more rapid diffusion of ions in the ordered mesoporous channels of CS-2, regardless of its smaller mesopore size than that of

![Figure 4](https://example.com/figure4.png)

**Figure 4.** Cyclic voltammograms for (a) CS-1 and (b) CS-2 in 6 M KOH at different voltage scan rates.

<table>
<thead>
<tr>
<th>TABLE 2: The Gravimetric Capacitance of CS-1 and CS-2 under Different Measurement Conditions in a Three-Electrode System</th>
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<tr>
<td>capacitor type sample</td>
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<td>CS-2</td>
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<tr>
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a The specific surface capacitance \(C_s\) was calculated by the equation \(C_s = C_g/S_{\text{BET}}\), where \(C_g\) is the gravimetric capacitance listed in Table 2 and \(S_{\text{BET}}\) is the BET specific surface area listed in Table 1.

![Figure 5](https://example.com/figure5.png)

**Figure 5.** The retained capacitance change of CS-1 and CS-2 with voltage scan rates.

the voltage sweep rates, it is still unable to precisely describe the actual electrochemical diffusion process. Hence, it is quite important to further investigate the influence of ordered mesoporous structure on ion diffusion based on EIS, which has been considered as a powerful method to obtain dynamic ion diffusion information.\textsuperscript{13–16}

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CS-1. Therefore, CS-2 shows a closer approach to pure capacitive behavior, which means that the ions are able to access more electrochemically active mesoporous surfaces in CS-2 than in CS-1 at the same AC frequency in the medium region.

3.4. Impact of Pore Packing Defects on Ion Transport. Both the CV and EIS methods clearly demonstrate that CS-2 exhibits a superior capacitive performance to CS-1, which means that the mesoporous structure of CS-2 favors ion diffusion. To account for the inferior mesopore size of CS-2 to that of CS-1, the intense negative impact of PPDs on ion diffusion must be clarified.

Impedance behavior on porous electrodes can be understood by ion penetrability: at higher penetrability most of the electrode surface should be detected, while at lower penetrability, only part of the porous electrode surface would be detected. On this basis, CS-2 should possess a higher penetrability than CS-1 does, as discussed previously.

The ion penetrability $\alpha$ can be expressed by the following formula:

$$\alpha = \frac{1}{2l_p} \sqrt{\frac{r}{C_d \omega R}} \quad (1)$$

where $r$ is the pore radius, $l_p$ is the pore length, $R$ is the inner-pore electrolyte resistance, $C_d$ is the specific surface EDL capacitance at the interface between the carbon phase and the electrolytes, and $\omega$ is the angular frequency.

It can be seen that the value of penetrability is directly proportional to $r^{1/2}$, $l_p^{-1}$, and $R^{-1/2}$. As the mesopore diameter for CS-1 is 5.5 nm, larger than that of CS-2, the values of $l_p^{-1}$ and $R^{-1/2}$ for CS-1 should be much smaller than those of CS-2, which would, in turn, give a lower penetrability value for CS-1 in order to be consistent with its poor capacitive behavior as determined by CV and EIS.

As observed from TEM images, the morphology of 2-D OMCs was rodlike particles with length at the micrometer scale and the primary mesopores running through the OMC rods. Therefore, it should be possible to estimate the primary mesopore length by evaluating the OMC rod length. From the OMC rod length distribution illustrated in Figure 7, CS-1 and CS-2 possess a similar rod length distribution with their peak values located between 0.8 and 1.2 $\mu$m, which indicates a similar primary mesopore length and hence a similar $l_p^{-1}$ value for both CS-1 and CS-2.

It has been deduced that the penetrability for CS-1 could be smaller than that for CS-2 only in the case of a smaller value of $l_p^{-1}$ and $R^{-1/2}$ for CS-1 than for CS-2. However, the $l_p^{-1}$ value has been proved to be similar for both CS-1 and CS-2. Consequently, the value of $R^{-1/2}$ for CS-1 must be greatly smaller than that for CS-2. As a result, the electrolyte ion resistance, $R$, in the mesoporous channels of CS-1 must be much larger than that of CS-2. The large ion diffusion resistance $R$ must have had a serious negative effect on reducing the ion diffusion ability, which, in turn, would have lowered the ion penetrability for CS-1.

As determined by TEM and SAXRD, CS-1 is believed to have possessed much more pore packing defects than CS-2. In other words, the PPDs for CS-1 are greater than that for CS-2 and, hence, will contribute to a large inner-pore electrolyte resistance, as deduced above.
Effect of Pore Packing Defects on Ionic Transport

When electrolyte ions transport into a pore under the stimulation of an AC signal, the ions will penetrate deeper along the porous channel with decreasing frequency until they have accessed the entire pore surface.\(^{14,15}\) Unfortunately, ion diffusion is intensely dependent on the actual pore channel conditions rather than an ideally cylindrical pore without any defects.\(^{14,15}\) The distributed pore packing defects along the mesoporous channels will constrict the mesopores to some extent. When the PPDs are severe and lead to a confined effect on pore diameters, as pointed out by the arrows in Figure 1b, the electrolyte ions will be restricted in the narrowed parts of the channels, and will transport more slowly than in the regular defect free mesopores (Figure 1d). This phenomenon would certainly increase the transport resistance in the mesoporous channels and hence result in a decrement of the ion penetrability, as observed for CS-1. Therefore, the capacitive behavior of CS-1 at high voltage scan rates was inferior to that for CS-2. Evidently, for 2-D hexagonal OMCs with similar PSD, the dynamic ion diffusion behavior in mesoporous channels will be strongly controlled by pore packing defects rather than by pore size distributions.

4. Conclusion

The capacitive behaviors of two different 2-D hexagonal OMCs were investigated based on CV and EIS. It was found that for an OMC with a larger mesopore size the capacitive behavior was inferior to that of an OMC with a smaller mesopore size. The reason is believed to lie in the difference between their ordered mesoporous structures. Pore packing defects have been proposed as the basis for interpreting these unexpected results. It is considered that for 2-D OMCs with similar mesopore diameters, the pore packing defects will determine the ion diffusion process in ordered mesopores at high current intensity. It is believed that a defect-free stable ordered mesoporous system will enable the electrolyte ion to transport more efficiently and will ultimately enhance the total EDLC system performance.

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Supporting Information Available: Electrochemical measurement results in a two-electrode system and TG/DSC results, including the TG curves of silica templates (Figure 1S), the DSC curves of OMCs (Figure 2S), the gravimetric capacitance values in a two-electrode system (Table 1S), and the specific surface capacitance values in a two-electrode system (Table 2S). This material is available free of charge via the Internet at http://pubs.acs.org.

References and Notes