Influence of Preparation Conditions on Structural Stability of Ordered Mesoporous Carbons Synthesized by Evaporation-induced Triconstituent Co-assembly Method

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Various ordered mesoporous carbons (OMCs) have been prepared by evaporation-induced triconstituent co-assembly method. Their mesostructural stability under different carbon content, aging time and acidity were conveniently monitored by X-ray diffraction, transmission electron microscopy, and \textsuperscript{N}_2 sorption isotherms techniques. The results show mesostructural stability of OMCs is enhanced as the carbon content increases from 36\% to 46\%, further increasing carbon content deteriorates the mesostructural stability. Increasing aging time from 0.5 h to 5.0 h make the mesostructural stability go through an optimum (2.0 h) and gradually reduce framework shrinkage of the OMCs. Highly OMCs can only be obtained in the acidity range of 0.2 – 1.2 mol/L HCl, when the acidity is near the isoelectric point of silica, the resulting OMCs have the best mesostructure stability. Under the optimum condition, the carbon content of 46\%, aging time of 2.0 h, and 0.2 mol/L HCl, the resulting OMCs have the best mesostructure stability and the highest BET surface areas of 2281 m$^2$/g.

Key words: Triblock copolymer, Mesostructural stability, Self-assembly, Ordered mesoporous carbon

I. INTRODUCTION

Ordered mesoporous carbon (OMC), one of the most promising mesoporous materials [1–3], possesses high specific surface areas, large uniform pores with narrow distribution and regular frameworks, making it more and more attractive for a number of applications [4, 5]. In these regards, significant efforts have been devoted to preparing ordered OMCs [6, 7].

The nanocasting method, also called hard-template method, has been used extensively in the preparation of mesoporous carbons [8]. However, extra step was needed to prepare ordered templates which make it costly and fussy [9]. An alternative method for the preparation of OMCs is soft-template self-assembly method [10, 11]. The essence of such method is the direct use of the surfactant as templates for generation of porous carbon structures without the extra step of generating templating silica structures [12]. Block copolymers, especially the Pluronic block copolymers (such as P123, F127), are used by many groups [13, 14] because of their commercially availability and inexpensiveness. However, structural shrinkage during high-temperature carbonization process leads to low specific surface areas and low pore volumes for OMCs. The triconstituent self-assembly strategy as one of the soft-template self-assembly methods is an effective solution for reducing framework shrinkage [15]. Zhao’s group demonstrated an evaporation-induced triconstituent copolymerization approach to prepare ordered mesoporous carbon-silica nanocomposites with “reinforced concrete” structured frameworks (silica is “reinforcement”, carbon is “concrete”) and large mesopore OMCs [16]. The presence of rigid silicates can not only greatly reduce skeleton shrinkage during the high-temperature carbonization but also can increase the specific surface areas and pore volumes of OMCs after silicas are removed. While, this makes the required reaction conditions stringent because of the complicated competition and cooperation among the inorganic-inorganic, inorganic-organic, and organic-organic interactions [16]. It has been shown that many factors such as reaction temperature, acidity, solvent and salt effect can influence the self-assembly process and the mesostructure stability of the resulting OMCs [13, 16].

In spite of the researches on the mesostructure stability of OMCs under various preparation conditions, these were principally limited to the evaluation of the stability of mesoporous materials prepared by dual-constituent self-assembly. The mesostructure stability of OMCs prepared by triconstituent evaporation-induced co-asse-
mbly method has little been reported.

In this work, we synthesized variable OMCs through solvent evaporation-induced tricomponent self-assembly method (EISA) using preformed resol as an organic precursor, prehydrolyzed TEOS as an inorganic precursor, and triblock copolymer F127 as a soft-template. More importantly, the mesostructural stability of prepared OMCs has been systematically investigated by varying carbon content, aging time and acidity. For the first time, we observed that increasing aging time from 0.5 h to 5.0 h can gradually reduce framework shrinkage of the OMCs and make the mesostructural stability go through an optimum. Appropriate carbon content and acidity can improve the mesostructural stability of the OMCs. The resulting OMCs prepared under the optimal conditions have high BET surface areas, large pore volumes and large mesopores.

II. EXPERIMENTS

A. Materials

Poly(propylene oxide)-block-poly(ethylene oxide)-block-poly(propylene oxide) triblock copolymer Pluronic F127 (PEO$_{106}$-PPO$_{70}$-PEO$_{106}$) was purchased from Sigma-Aldrich Corp. Tetraethyl orthosilicate (TEOS), phenol, formalin solution (37%), NaOH, HCl, and HF solution (40%) and ethanol were purchased from Sinopharm Chemical Reagent Co., Ltd.

B. Synthesis of ordered mesoporous carbon-silica nanocomposites and OMCs

Mesoporous carbon-silica nanocomposites were prepared by tricomponent co-assembly of resol, oligomer silicates from TEOS, and triblock copolymer F127 template according to the modified literature method [16]. In a typical preparation, 1.6 g of block copolymer F127 and 1.0 mL of 0.2 mol/L HCl were dissolved in 10.0 mL ethanol and stirred for 1.0 h at 40 °C. Next, 2.08 g of TEOS and 5.0 g of 20% resol’s ethanol solution [13] were added in sequence. After stirring for 2.0 h, the mixture was transferred into dishes. It took 7.0 h at room temperature to evaporate ethanol and 24 h at 100 °C in an oven to thermopolymerize. Then, the products were scraped from the dishes and ground into fine powders. Calcination was carried out in a tubular furnace at 850 °C for 5.0 h under N$_2$ flow to get ordered mesoporous carbon-silica nanocomposites, named as 0.2-OM-CS-46-2.0. “z-OM-CS-x-y” denotes the ordered mesoporous carbon-silica nanocomposite samples, wherein z represents the percentage of the carbon-compound content in the carbon-silica nanocomposite after 850 °C calcinations, which was calculated based on the mass loss of the carbon-silica nanocomposites before and after calcination in air, y represents the aging time, z represents the HCl concentration.

After carbon-silica nanocomposites were immersed in 10% HF solutions for 24 h, silicas were removed and mesoporous carbons were left named as “z-OM-C-x-y”. We can obtain various OMCs by easily varying carbon content, aging time and HCl concentration. The typical samples under different preparation conditions are listed in Table I.

C. Characterization

The small angle X-ray diffraction (XRD) measurements were taken on a TTR-III (Rigadu, Japan) using Cu Kα radiation (40 kV, 200 mA). Nitrogen sorption isotherms were measured at 77 K with a Micrometrics ASAP 2020 analyzer (Micromeritics, USA). Transmission electron microscopy (TEM) measurements were taken on a JEOL 1010 microscope (JEOL, Japan) operated at 200 kV.

III. RESULTS AND DISCUSSION

A. Ordered mesoporous carbon-silica nanocomposites and OMCs

We take the mesoporous carbon-silica nanocomposite 0.2-OM-CS-46-2.0 and corresponding OMC 0.2-OM-C-46-2.0 as examples. The small angle XRD patterns (Fig.1) for both show three diffraction peaks at a 2θ range of 0.5°–2° that can be indexed as (10), (11), and (20) reflections associated with 2D hexagonal $P6_3mm$ symmetry [13]. This observation indicates that both have a high degree of hexagonal mesoscopic organization. The intense (10) peaks reflect d-spacings of 9.3 and 9.8 nm, which correspond to large unit-cell parameters (Table II), those are much larger than FDU-15 ($a_0=8.6$ nm) [13]. These results indicate that the presence of silica in the nanocomposite can efficiently reduce

<table>
<thead>
<tr>
<th>Sample</th>
<th>$R$/g</th>
<th>F127/g</th>
<th>y/h</th>
<th>z/(mol/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2-OM-CS-46-2.0</td>
<td>5.0</td>
<td>1.6</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>0.2-OM-C-36-2.0</td>
<td>2.5</td>
<td>1.0</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>0.2-OM-C-46-2.0</td>
<td>5.0</td>
<td>1.6</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>0.2-OM-C-61-2.0</td>
<td>10.0</td>
<td>2.3</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>0.2-OM-C-46-0.5</td>
<td>5.0</td>
<td>1.6</td>
<td>0.5</td>
<td>2.0</td>
</tr>
<tr>
<td>0.2-OM-C-46-1.0</td>
<td>5.0</td>
<td>1.6</td>
<td>1.0</td>
<td>2.0</td>
</tr>
<tr>
<td>0.2-OM-C-46-5.0</td>
<td>5.0</td>
<td>1.6</td>
<td>5.0</td>
<td>2.0</td>
</tr>
<tr>
<td>0.0-OM-C-46-2.0</td>
<td>5.0</td>
<td>1.6</td>
<td>2.0</td>
<td>0.0</td>
</tr>
<tr>
<td>1.2-OM-C-46-2.0</td>
<td>5.0</td>
<td>1.6</td>
<td>2.0</td>
<td>1.2</td>
</tr>
<tr>
<td>2.0-OM-C-46-2.0</td>
<td>5.0</td>
<td>1.6</td>
<td>2.0</td>
<td>2.0</td>
</tr>
</tbody>
</table>

$R$ is resol solution.
TABLE II Physicochemical properties of the ordered mesoporous carbon-silica nanocomposites and of the corresponding OMCs.

<table>
<thead>
<tr>
<th>Sample</th>
<th>(a_0/\text{nm})</th>
<th>(D/\text{nm})</th>
<th>(S_{\text{BET}}/(\text{m}^2/\text{g}))</th>
<th>(V_t/(\text{cm}^3/\text{g}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2-OM-CS-46-2.0</td>
<td>10.7</td>
<td>6.5</td>
<td>455</td>
<td>0.55</td>
</tr>
<tr>
<td>0.2-OM-C-36-2.0</td>
<td>11.2</td>
<td>4.0</td>
<td>1560</td>
<td>1.49</td>
</tr>
<tr>
<td>0.2-OM-C-46-2.0</td>
<td>11.3</td>
<td>6.5</td>
<td>2281</td>
<td>1.92</td>
</tr>
<tr>
<td>0.2-OM-C-61-2.0</td>
<td>10.1</td>
<td>3.6</td>
<td>813</td>
<td>0.24</td>
</tr>
<tr>
<td>0.2-OM-C-46-0.5</td>
<td>9.7</td>
<td>5.4</td>
<td>1123</td>
<td>1.55</td>
</tr>
<tr>
<td>0.2-OM-C-46-1.0</td>
<td>10.0</td>
<td>6.4</td>
<td>1346</td>
<td>1.92</td>
</tr>
<tr>
<td>0.2-OM-C-46-5.0</td>
<td>11.4</td>
<td>4.7</td>
<td>1179</td>
<td>1.54</td>
</tr>
<tr>
<td>1.2-OM-C-46-2.0</td>
<td>10.9</td>
<td>6.3/2.5</td>
<td>1833</td>
<td>1.94</td>
</tr>
<tr>
<td>2.0-OM-C-46-2.0</td>
<td>9.9</td>
<td>3.9</td>
<td>385</td>
<td>0.42</td>
</tr>
</tbody>
</table>

\(a_0\), the cell parameter, was calculated by using formula \(a_0=2d/\sqrt{3}\). \(D\) is the pore diameter. \(S_{\text{BET}}\) is the BET surface area. \(V_t\) is the total pore volume.

The \(\text{N}_2\) sorption isotherms (Fig.3(a)) of the two samples yield type-IV curves with sharp capillary condensation steps in the relative pressure range from 0.6 to 0.7 and obvious \(H_1\)-type hysteresis loops which are typical of mesoporous materials with cylindrical channel [16]. The BET surface area and the total pore volume of the two samples are calculated to be 455, 2281 \(\text{m}^2/\text{g}\) and 0.55, 1.92 \(\text{cm}^3/\text{g}\) (Table II), respectively. The increased BET surface area for 0.2-OM-C-46-2.0 is ascribed from plenty of small pores inside carbon walls after removal of silica. The pore diameters are all about 6.5 nm with a narrow distribution (Fig.3(b)), which are larger than the FDU-15 [13], indicative of a smaller framework shrinkage than the latter case.

B. Carbon content

Carbon contents in the nanocomposites not only impact the mesostructure regularity of nanocomposites [16], but also influence the mesostructure stability of the corresponding OMCs. Song’s group reported that if the carbon content is more than 90%, the polymer-silica composite films contain disordered small pores and exhibit low porosity [17]. With loadings of the carbon content below 68%, the films are formed well-ordered and highly resolved near hexagonal mesostructure [17], while it is rarely reported how carbon contents impact the mesostructural stability of OMCs from variable carbon-silica nanocomposite powders. Therefore, we systematically studied mesostructural stability of the carbon-silica nanocomposite powders and the corresponding OMCs by varying the carbon content. The carbon content can vary from zero to infinity [16]. We used 36%, 46%, and 61% of carbon contents as exam-

FIG. 1 Small-angle XRD patterns of the mesoporous carbon-silica nanocomposite 0.2-OM-CS-46-2.0 and corresponding OMC 0.2-OM-C-46-2.0.

FIG. 2 TEM images of the ordered mesoporous carbon-silica nanocomposites 0.2-OM-CS-46-2.0 ((a) and (c)) and corresponding OMC 0.2-OM-C-46-2.0 ((b) and (d)), viewed from the [110] ((a) and (b)) and [001] ((c) and (d)) directions. The insets are the corresponding fast Fourier transform (FFT) diffractography.

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FIG. 3 
N\textsubscript{2} sorption isotherms (a) and pore size distribution (b) of the ordered mesoporous carbon-silica nanocomposites 0.2-OM-CS-46-2.0 and corresponding OMC 0.2-OM-C-46-2.0.

FIG. 4 Small-angle XRD patterns of (a) the mesoporous carbon-silica nanocomposites with different carbon content and (b) the corresponding OMCs after removal of silica.

The small-angle XRD patterns of the carbon-silica nanocomposites with different carbon content are illustrated in Fig.4(a). All nanocomposites exhibit one intense and two weak diffraction peaks. Those can be indexed as (10), (11), and (20) reflections, suggesting that all nanocomposites have highly ordered mesostructure. With an increase of carbon contents, the cell parameters decrease (Table II) and the (10) diffraction peak becomes wide, suggesting that the asymmetric framework shrinkage increase and the mesostructure stability deteriorate gradually. While after removal of silica, the resulting OMCs frameworks exhibit different phenomenon from the mother nanocomposites. The small-angle XRD patterns of all OMCs are shown Fig.4(b). The (10) diffraction peaks of all the OMCs become wider compared to corresponding mother nanocomposites, implying that the mesostructure regularity of OMCs reduces to some extent, while the 0.2-OM-C-46-2.0 shows the sharpest (10) diffraction peaks, suggesting its best mesostructure stability in all of the OMCs. That means the mesostructure stability of OMCs goes through an optimum as the carbon content increases, which is different from the mother nanocomposites.

Similar phenomenon can be observed from TEM images (Fig.5). The carbon-silica nanocomposite 0.2-OM-CS-36-2.0 shows large domains of highly ordered stripe-like arranged images (Fig.5(a)), indicating highly ordered mesostructure. While the stripe-like arranged image of the 0.2-OM-CS-46-2.0 becomes slightly curving (Fig.5(b)). Further increasing carbon content to 61\%, the TEM image (Fig.5(c)) of 0.2-OM-CS-61-2.0 becomes more curving, implying the mesostructure are destroyed gradually. After removal of silica, the TEM images (Fig.5 (d)–(f)) of all the resulting OMCs show uneven surface. The 0.2-OM-C-36-2.0 shows many imperfect mesopore channels (marked with a circle in Fig.5(d)). When carbon content increases to 46\%, the 0.2-OM-C-46-2.0 exhibits a high-quality hexagonal mesostructure (Fig.5(e)). Further increasing carbon content, the mesostructure of the 0.2-OM-C-61-2.0...
2.0 are destroyed seriously (Fig. 5(f)). It is hypothesized that lower carbon content (higher silica content) can more effectively reduce framework shrinkage which makes the nanocomposites have better mesostructure stability, while removal of silica leads to many voids which destroy the carbon framework and make the mesostructure stability of the resulting OMCs worse. When further increasing carbon contents (decreasing silica content), the asymmetric framework shrinkage intensifies, which makes the mesostructure stability of the nanocomposites deteriorate gradually and further leads to bad mesostructure stability of the resulting OMCs. It can be concluded that the 46% of carbon content is the best condition for the mesostructure stability of OMCs.

For validating our hypothesis, the N\textsubscript{2} sorption techniques are adopted. The N\textsubscript{2} sorption isotherms and the pore diameter distributions of all OMCs derived from the corresponding carbon-silica nanocomposites are shown in Fig. 6. The 0.2-OM-C-36-2.0 yields a type-IV curve with a sharp capillary condensation step in the relative pressure ranged from 0.4 to 0.8 and an obvious H\textsubscript{2}-type hysteresis loop with a large space at high relative pressure, reflecting a large calculated pore size caused by the resulting voids after removal of silica inside carbon pore walls. Simultaneously, the wide pore distributions indicate the mesostructure is damaged to some extent. Its surface areas and total pore volume are 1560 m\textsuperscript{2}/g and 1.49 cm\textsuperscript{3}/g, respectively (Table II). The 0.2-OM-C-46-2.0 also shows a type-IV curve but the capillary condensation step becomes more sharper and shifts to higher relative pressure (Fig. 6(a)) compared to 0.2-OM-C-36-2.0, suggesting that larger pores with a narrower pore size distribution (Fig. 6(b)). Another distinguished phenomenon is extremely high BET surface areas of 2281 m\textsuperscript{2}/g which also imply the better mesostructure stability of the 0.2-OM-C-46-2.0. While further increasing the carbon content, the 0.2-OM-C-61-2.0 shows a type-IV curve with the widest pore distribution. Its BET surface area is the lowest (813 m\textsuperscript{2}/g) among the three OMCs ascribed from the asymmetric framework shrinkage during the high temperature calcination, indicating the worst mesostructure. The 46% of carbon content makes the OMCs have the best mesostructural stability and highest BET surface area. All the results are consistent with the XRD and TEM observations.

C. Aging time

On the basis of sol-gel chemistry, the aging time of TEOS under HCl acid-catalyzed condition affects remarkably the polymerization and cross-linking degree of silicates which is vital for triconstituent self-assembly [16, 18]. Sanchez and co-workers reported that the polymerization degree of inorganic precursors should be low enough to form a moldable inorganic-organic framework at the initial assembly stage of inorganic species with organic surfactant [18]. However, the understanding of the mechanism is limited. Herein, we studied the mesostructural stability of ordered mesoporous carbon-silica nanocomposites and the corresponding OMCs (using 0.2-OM-CS-46-y series as examples, the preparation conditions are listed in Table I) with various aging time ranged from 0.5 h to 5.0 h.

The small angle XRD patterns of the nanocomposites 0.2-OM-CS-46-y (Fig. 7(a)) reveal that the diffraction peaks of (10), (11), and (20) became sharper and stronger along with increasing aging time, suggesting the mesostructure regularity is improved gradually. The (10) peaks shift to lower angle implying that the cell parameter increases from 9.7 nm to 11.2 nm. This phenomenon clearly demonstrates that increasing aging...
time from 0.5 h to 5.0 h can efficiently reduce framework shrinkage of nanocomposites. After removal of silica, the resulting OMCs show similar phenomenon, but their mesostructural stability goes through an optimum based on the small angle XRD observations (Fig.7(b)). We propose that for the carbon-silica nanocomposites, prolonging the aging time from 0.5 h to 5.0 h increases the polymerization and cross-linking degree of silicates. Then large domain “reinforcement” silica aggregations in carbon walls can be obtained [16], which can efficiently reduce the asymmetric framework shrinkage and improve the mesostructural stability of the carbon-silica nanocomposites. However, when further prolonging the aging time, phase separation occurs. For the corresponding OMCs, the voids interpenetrated with carbon walls become larger and larger with increasing aging time after etching silica, which result in uneven surface, poor mechanical intensity and insufficient mesostructural stability. Therefore, there exists an optimal aging time, 2.0 h, which makes the 0.2-OM-46-y samples have the best mesostructure stability.

N₂ sorption isotherms (Fig.8(a)) of 0.2-OM-C-46-y reflect high uniformity of mesopore sizes. The pore size distribution curves (Fig.8(b)) show the primary pores and pore size distributions become large and narrow respectively with increasing aging time from 0.5 h to 2.0 h, implying the mesostructure stability of 0.2-OM-C-46-y is improved. While, when aging time prolong to 5.0 h, the pore size and the pore size distribution of the 0.2-OM-C-46-5.0 become small and wide, suggesting its poor mesostructure. The results show an optimum of 2.0 h makes the 0.2-OM-C-46-y have the best mesostructure stability, which is in accordance with

D. Acidity

Another crucial factor that influences the self-assembly and mesostructure is acidity. It’s well-known that by increasing acidity, the protonation of the EO moieties of F127 will increase their hydrophilicity, thus leading to the structural transform [19]. Additionally, increasing acidity will accelerate the polymerization and cross-linking degree of silicates, then further impact the competition and balance of triconstituent. But how acidity impact the mesostructure is unclear because of the complexity of triconstituent self-assembly.

To assess the effect of acidity on the mesostructure of nanocomposites and OMCs (using z-OM-C(CS)-46-2.0 as examples, the preparation conditions are listed in Table I), small angle XRD has been employed as illustrated in Fig.9. Here, it should be pointed out that F127 is unsolvable under basic condition in our process. Under neutral conditions (HCl content is 0.0 mol/L), the XRD pattern of 0.0-OM-CS-46-2.0 nanocomposite (Fig.9(a)) exhibits the widest (10) diffraction peak and the lowest signal to noise ratio, suggesting its disordered mesostructure. When HCl content increases to 0.2 mol/L, the 0.2-OM-CS-46-2.0 shows three strong diffraction peaks, implying the highly ordering. Further increasing HCl content, the diffraction peaks become weak gradually, suggesting the mesostructure stability deteriorates by degrees. Meanwhile, the (10) peaks shift to high angle (implying the cell parameter decrease) even if increasing HCl content improves the polymerization and cross-linking degree of silicates that can efficiently reduce the asymmetric framework shrinkage, which is inverse to that of aging time. The corresponding OMCs exhibit the similar phenomenon (Fig.9(b)). It can be concluded that highly OMCs can
only be obtained in the acidity range of 0.2−1.2 mol/L HCl. When the HCl concentration is 0.2 mol/L, the resulting OMCs have the best mesostructural stability. We suggest that the 0.2 mol/L HCl concentration (in our system, pH=2.0), which is near the isoelectric point of silica (pH=2.0), makes the oligomer silicates from TEOS have the highest silanol concentration. Therefore the hydrogen bonding interaction between the triconstituent is enhanced, which results in highly ordered mesostructure. When acidity is too low (such as 0.0 mol/L HCl) or too high (such as 2.0 mol/L HCl), the decreased silanol density of oligomer silicates weakens the hydrogen bonding interaction, which leads to decreased mesostructure [20]. Meanwhile, higher HCl concentration greatly improves the polymerization and cross-linking degree of silicates, which destroy the completion and balance of triconstituent and further induce to “heterogenous” frameworks that intensify asymmetric framework shrinkage during calcination. In addition, higher HCl concentration increases the ion intensity of the system, reduces the hydrophilicity of the PEO moieties of F127 which is similar to salt effect [21], then also weakens the hydrogen bonding interaction between triconstituent and finally leads to poor mesostructure of the carbon-silica nanocomposites and corresponding OMCs.

$N_2$ sorption isotherms (Fig. 10(a)) of z-OM-C-46-2.0 further validate our hypothesis. 0.2-OM-C-46-2.0 exhibits a typical IV curve with a $H_1$ hysteresis loop and a narrow pore size distribution (Fig.10(b)), implying its high ordering mesostructure. Its BET surface areas and total pore volume are 2281 m$^2$/g and 1.92 cm$^3$/g, respectively (Table II). However, for 1.2-OM-C-46-2.0 sample, the $H_1$ hysteresis loop shifts to low relative pressure compared to that of 0.2-OM-C-46-2.0, suggesting smaller pore diameter (Fig.10(b)) and Table II). Its BET surface areas decrease to 1833 m$^2$/g. The detective bimodal-pores (Fig.10(b)) also imply the increased polymerization and cross-linking degree of silicates [16]. It is noticeable that for 2.0-OM-C-46-2.0 the mesostructure has been essentially destroyed and the porosity is very low as reflected from the isotherm, which is in agreement with the XRD result (Fig.9(b)). The widest pore size distribution (Fig.10(b)) and the lowest specific surface areas (385 m$^2$/g) also reflect its poor ordering of mesostructure. Therefore, we conclude that the highly ordered mesoporous carbon materials can only be obtained in an acidity range of 0.2−1.2 mol/L HCl. When the acidity (HCl concentration) is near the isoelectric point of silica (0.2 mol/L), the resulting OMCs have the best mesostructure stability.

IV. CONCLUSION

We systematically investigate the mesostructural stability of the prepared OMCs through evaporation-induced triconstituent co-assembly method by varying carbon content, aging time, and acidity. The results show that mesostructural stability of OMCs is enhanced as the concentration of carbon increases from 36% to 46%, further increasing carbon content destroys their mesostructure. Framework shrinkage of the OMCs can be gradually reduced with increasing aging time from 0.5 h to 5.0 h. While after etching silica, large voids are formed, which destroy the carbon framework and result in poor mesostructure of OMCs. The 2 h of aging time makes the OMC have the best ordering; Highly OMCs can only be obtained in the acidity range of 0.2−1.2 mol/L HCl. When the acidity (0.2 mol/L HCl) is near the isoelectric point of silica, the highest silanol density of oligomer silicates from TEOS makes the triconstituent have the strongest hydrogen-bonding interaction and results in highly OMCs. The 46% of carbon content, 2 h of aging time and 0.2 mol/L HCl concentration make the resulting OMCs have the best mesostructure stability and the highest BET surface areas of 2281 m$^2$/g, which make them promising candidates for applications in energy storage and sorption.

V. ACKNOWLEDGMENT

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