



## Preparation of ordered mesoporous carbons with ammonia modification for Orange II adsorption

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### ABSTRACT

In this study, the ordered mesoporous carbon (CMK-3) was prepared using SBA-15 as hard-template and furfuryl alcohol as the carbon source and then ammonia modification via tube furnace under different temperatures. The effect of ammonia activation temperature and the sorption behavior of Orange II (O II) on ordered mesoporous carbons were investigated. The textural property and surface chemistry of the ordered mesoporous carbons also were investigated by N<sub>2</sub> adsorption, elemental analysis, and X-ray Photoelectron Spectroscopy. Results indicated that the formation of mesopores volume, specific surface area, total pore volume, and the average pore diameter could be enhanced with ammonia modification for the mesoporous carbon. Furthermore, the nitrogen content of as-prepared samples increased with increasing modification temperature. In addition, the results indicated that the mesoporous carbon CMK-3-1173 treated at 900°C has developed specific surface area and abundant mesopores, which showed the largest adsorption capacity for O II among all samples investigated.

*Keywords:* CMK-3; Ammonia modification; Nitrogen functional group; Adsorption

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### 1. Introduction

Activated carbons have a high degree of porosity and an extensive surface, and are commonly utilized as adsorbents for the removal of color, odor, organic or inorganic matter, and waste effluents from aqueous solutions [1]. The prominent adsorption performance of activated carbon for organic pollutants is attributed to its abundant pore structure and large specific surface area. However, the domination of micropores in common activated carbons hinders its applications particularly for the adsorption of the large molecule or macromolecule.

Mesoporous (2–50 nm) carbon which has an open pore structure and a large pore size provides marked advantages in bulky molecular or macromolecule adsorption [2]. There are mainly two ways to prepare mesoporous carbon materials. One is preparing mesoporous activated carbons from carbon precursors, such as bamboo [3] and waste tires [4]. The other one of the most extensively studied areas in materials chemistry is the fabrication of mesoporous materials through template approaches. Han et al. [5] studied the adsorption of bulky dyes onto the mesoporous carbon prepared using silica nanoparticles as templates, founding that the adsorption capacities of mesoporous carbon for bulky molecule dyes were much higher than that of commercial activated carbon.

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The surface chemistry and the pore structure of porous carbon determine its application. The functional groups of surface have significant influence on the surface chemistry of porous carbons. There are many methods for the surface modification of activated carbon including acid treatment, heat treatment, ammonia treatment, microwave treatment, and so on [6,7]. Faria et al. [8] concluded that basic carbons were the most efficient for the removal of anionic dyes. Mangum et al. [9] modified activated carbon fiber with dry ammonia and showed the reactivity of ammonia gas with carbon surface and consequently the presence of N-containing functionalities on the activated carbon fiber surface.

Synthetic dye is used for the coloration of various substrates, such as paper, leather, food, cosmetics, textiles, and so on [10].  $7 \times 10^5$  tons of synthetic dye is annually discharged into an aquatic ecosystem [11]. The discharge of dye effluents into the environment is currently one of the world's major environmental problems for the dramatic decrease of photosynthetic activity of aquatic lives due to reduced penetration of sunlight. Furthermore, most of the dyes are toxic and even carcinogenic, which pose a serious hazard to aquatic living organisms [12].

In this study, ordered mesoporous carbon CMK-3 was prepared using SBA-15 silica as hard-template and furfuryl alcohol as the carbon source and then modified by ammonia via a tube furnace so as to introduce basic nitrogen functionalities onto the CMK-3 surface. The effect of the different ammonia modification temperatures on the mesoporous carbon structural property was investigated. In order to elucidate the adsorption mechanism, the material surface chemistry characterization of CMK-3 before and after modification was carried out and the dye adsorption capacity of mesoporous carbon was also tested.

## 2. Experimental

### 2.1. Materials

Orange II (O II), Triblock copolymer  $\text{EO}_{20}\text{PO}_{70}\text{EO}_{20}$  (Pluronic P123, Aldrich), tetraethyl orthosilicate (TEOS, 98%, Aldrich), furfuryl alcohol, HCl, NaOH, and  $\text{H}_2\text{SO}_4$  were supplied from Sigma–Aldrich. All other reagents were of analytical grade.

### 2.2. Synthesis of mesoporous carbon CMK-3

The CMK-3 samples were synthesized by a nano-casting process using SBA-15 silica as template and furfuryl alcohol as the carbon source following

the synthesis procedure outlined by Jun et al. with slight revisions [13]. The SBA-15 template was prepared using the triblock copolymer,  $\text{EO}_{20}\text{PO}_{70}\text{EO}_{20}$  (Pluronic P123, Aldrich) as the template and TEOS (98%, Aldrich) as the silica source. The triblock copolymer Pluronic P123 was dissolved in hydrochloride solution at 35°C. Then TEOS was added to the mixture under magnetic stirring. The molar ratio of the final mixture was TEOS: P123: HCl:  $\text{H}_2\text{O}$  = 1: 0.0165: 5.755: 191.61. After the solution was magnetically stirred at 40°C for 24 h, it was transferred into an autoclave, then the sample aged in a steady state for 24 h at 100°C. The final product was filtrated, dried, and washed with distilled water and then calcined at 550°C in air at a ramping rate of 1°C/min and kept the temperature for 12 h to remove the organic template P123.

The CMK-3 samples were synthesized according to the following process. Briefly, 1 g of SBA-15 was mixed with 5 mL furfuryl alcohol by incipient wetness technique at room temperature under magnetic stirring for 24 h. The loaded sample was heated to 100°C to polymerize the furfuryl alcohol for 12 h and further carbonized at 160°C for another 12 h. Then, the resulting dark brown material was carbonized at 900°C (at a rate of 5°C/min) under nitrogen gas atmosphere and the state was maintained for 9 h to carbonize the polymer. Finally, the silica/carbon composite was filtered and then washed with 1 mol/L NaOH (50 vol.% ethanol + 50 vol.%  $\text{H}_2\text{O}$ ) at 100°C twice to remove the silica, and subsequently dried at 100°C for 12 h [14]. The CMK-3 products were obtained.

### 2.3. Modification of mesoporous carbon CMK-3

Heat treatment with ammonia gas at high temperatures was a simple and effective post-modification method to incorporate basic nitrogen-containing functional groups onto the surface of CMK-3. In this paper, ammonia treatment was conducted via tube furnace under different high temperatures, the nitrogen-containing functional groups generated from the ammonia treatment was loaded into CMK-3. The process is conducted with a homemade thermal treatment system as schematically shown in Fig. 1.

At first, 0.5 g of CMK-3 was placed in a quartz reactor and then high purity nitrogen was introduced to expel air. The sample was heated to 200°C at a heating rate of 5°C/min for 3 h under nitrogen in order to remove moisture and impurities. Afterwards, the furnace was heated to the desired temperature in nitrogen atmosphere, and then the gas was switched to ammonia gas. The sample was subsequently treated with ammonia (flow rate = 60  $\text{cm}^3/\text{min}$ ) for 80 min and

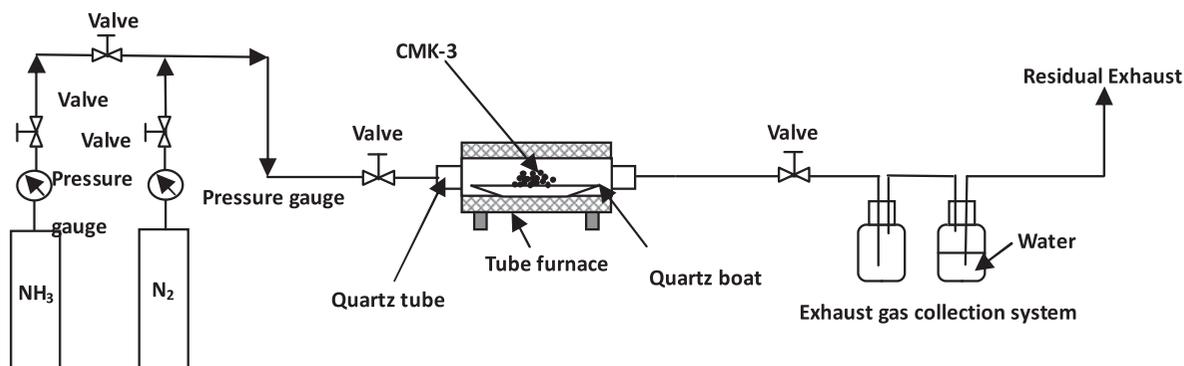


Fig. 1. Schematic diagram of Chemical vapor deposition (CVD) equipment.

then purged with nitrogen for 90 min, then cooling to room temperature under the same atmosphere. After that, the sample was washed using deionized water until the pH of the filtrate liquid reaches close to 7 and cooled overnight at room temperature. Finally, the sample was collected and stored in a desiccator. The nomenclature for the modified sample was based on modified reaction temperature. The sample was labeled as CMK-3-973K for that treated at 700 °C.

#### 2.4. Characterization of mesoporous carbon

Nitrogen adsorption isotherms were measured at 77 K using Micromeritics ASAP 2000 system. The specific surface area was calculated from the adsorption data in a relative pressure interval from 0.05 to 0.35 using the Brunauer–Emmett–Teller (BET) method. The pore size distribution curve was calculated by Barrett–Joyner–Halenda (BJH) method using the desorption branch of the isotherm. The t-plot method was employed to analyze the micropore volume and the BJH method was employed to estimate the mesopore volume. The total pore volume was calculated at the relative pressure of 0.995.

The low-angle X-ray powder diffraction patterns were recorded on a PW1830 (Philips) diffractometer using Cu K $\alpha$  radiation. The morphology and microstructures of the origin and modified CMK-3 were investigated using scanning electron microscopy (SEM, JEOL-6300) and transmission electron microscope (TEM, Philips CM20).

Elemental analysis was performed using Vario EL III Elemental Analyzer. A weighed sample was placed in a tin capsule and combusted at a high temperature (1,400 °C). Resultant reaction gas was then introduced into a series of adsorption column coupled with a thermal conductivity detection system. By measuring the difference in the electric conductivity between

helium reference and reaction gas, the element content of the sample was calculated.

##### 2.4.1. X-ray Photoelectron Spectroscopy (XPS)

XPS was adopted to determine the content of element C, N, O on the surface of CMK-3. XPS spectra of the elements were recorded on a Physical Electronics PHI 5600 XPS spectrophotometer with a monochromatic Al K $\alpha$  X-ray source. Atomic concentrations were calculated from the XPS survey spectra after correcting the relative peak areas by sensitivity factors.

#### 2.5. Dye adsorption

The adsorption behaviors of O II onto the origin CMK-3 and modified CMK-3 were measured according to the following procedure. Typically, 0.03 g adsorbent was added into a 50 mL O II aqueous solution (300 mg/L) and placed in a thermostat bath that is continuously shaken at 25 °C. All the adsorption experiments were carried out in triplicate. For dye adsorption experiment, a blank test was included (no adsorbent), which was used as reference to calculate the amount of dye removed. The concentration of upper solution ( $C_e$ ) was determined by UV/VIS Spectrophotometer (Lambda 20, Perkin Elmer) at 485 nm. The adsorption amount  $q_e$  (mg/g) at equilibrium was determined as follows:

$$q_e = \frac{(C_0 - C_e)V}{W} \quad (1)$$

where  $C_0$  (mg/L) and  $C_e$  (mg/L) are the liquid-phase equilibrium concentrations of O II at initial and equilibrium, respectively.  $V$  is the volume of the liquid solution (L), and  $W$  is the mass of the adsorbent used (g).

### 3. Results and discussion

#### 3.1. Structural and textural properties

The nitrogen adsorption–desorption curve provides valuable information for the adsorption mechanism and porous structure of the adsorbent. From the nitrogen adsorption–desorption isotherms of mesoporous carbon before and after modification, it can be seen that the obtained CMK-3 still have type IV isotherms after different ammonia modification temperatures the same as before, with a steep H1 hysteresis loop in the relative pressure range between 0.4 and 0.6, indicating the presence of framework mesopores with uniform size, which is a typical characteristic of mesoporous materials, the result also indicates that mesoporosity is preserved after modification [15]. Table 1 shows the change of pore structure before and after ammonia modification, it can be observed that the ammonia gas modification resulted in an obvious increase in surface area and total volume. Those after modification have larger BET surface area, pore volume and smaller amount of micropores in comparison with the original one. In addition, it is noteworthy that a definite increase is observed in the BET specific surface area and mesopore volume when modified reaction temperature increased from 700 to 900 °C. As can be seen in Table 1, the increase of the specific surface areas is from 1,181 to 1,746 m<sup>2</sup>/g, and the total volume is from 1.08 to 1.46 cm<sup>3</sup>/g. As seen in Fig. 2 and Table 1, the average pore size distribution of adsorbent also has been influenced by the heat modification temperature, the mesopous pore size of the CMK-3 samples slightly increased from 3.4 to 3.7 nm with increasing of modification temperature during ammonia tailoring, these changes can be attributed to the drastic conditions used in the temperature range of 700–900 °C, indicating that ammonia is acting as an activating agent. An increase in temperatures results in more modification thus increasing the pore size, specific areas, and total volume, but a decrease in the micropore volume was noted, which results from the thermal decomposition of the surface oxygen-containing groups that were either inside the micropores or blocking the micropore entrances. As

the increase in modification temperature would intensify a vigorous modification reaction on the surface of the adsorbent, it leads to carbon burn off and more micropores transforming into mesopores [16]. Additionally, it is noted that as the modification temperature is raised, there was slight shrinkage of the carbon matrix to offset the weight loss during charring resulting in a narrowing of the pores.

The effect of ammonia modification temperature on the mesoporous structure was investigated and the sample structure was measured by XRD and TEM. Fig. 3 shows the low-angle XRD patterns of original CMK-3 and modified CMK-3. The XRD patterns of CMK-3 and modified CMK-3 have three diffraction peaks, which can be indexed as (100), (110), and (200) reflections associated with hexagonal symmetry [17]. The presence of these three peaks in modified CMK-3 indicates that modified mesostructure has been retained and still has a high degree of hexagonal mesoscopic organization after ammonia modification. With modification, the diffraction peak of the three modified CMK-3 slightly moves to the left, indicating some change in the structure of the samples at high temperatures.

The TEM images of original and the ammonia-modified CMK-3 samples are shown in Fig. 4, indicating that the hexagonally ordered structure of modified CMK-3 is preserved after ammonia modification, and the pore size seems almost the same, which was in accordance with the XRD results.

As shown in Fig. 5, it is suggested that ammonia treatment under different temperatures could not make a drastic difference in the particle morphology. The particle morphology of the modified CMK-3 is similar to that of the original CMK-3, which consisted of many uniform rope-like domains.

The elemental analysis and XPS of original CMK-3 and modified CMK-3 are shown in Tables 2 and 3, respectively. From the elemental analysis, it can be observed that the carbon content of original CMK-3 is around 85.34% and there is little nitrogen present in the sample. It can be assumed that the increasing in nitrogen could be ascribed to the addition of some new nitrogen-containing groups to the sample surface,

Table 1  
Porous parameters of original and modified CMK-3

Sample	$S_{\text{BET}}$ (m <sup>2</sup> /g)	$V_{\text{total}}$ (cm <sup>3</sup> /g)	$V_{\text{mic}}$ (cm <sup>3</sup> /g)	$D_{\text{meso}}$ (nm)
CMK-3	1181.35	1.04	0.107	3.2
CMK-3-700	1294.84	1.09	0.037	3.4
CMK-3-800	1317.29	1.15	0.034	3.5
CMK-3-900	1746.09	1.46	0.006	3.7

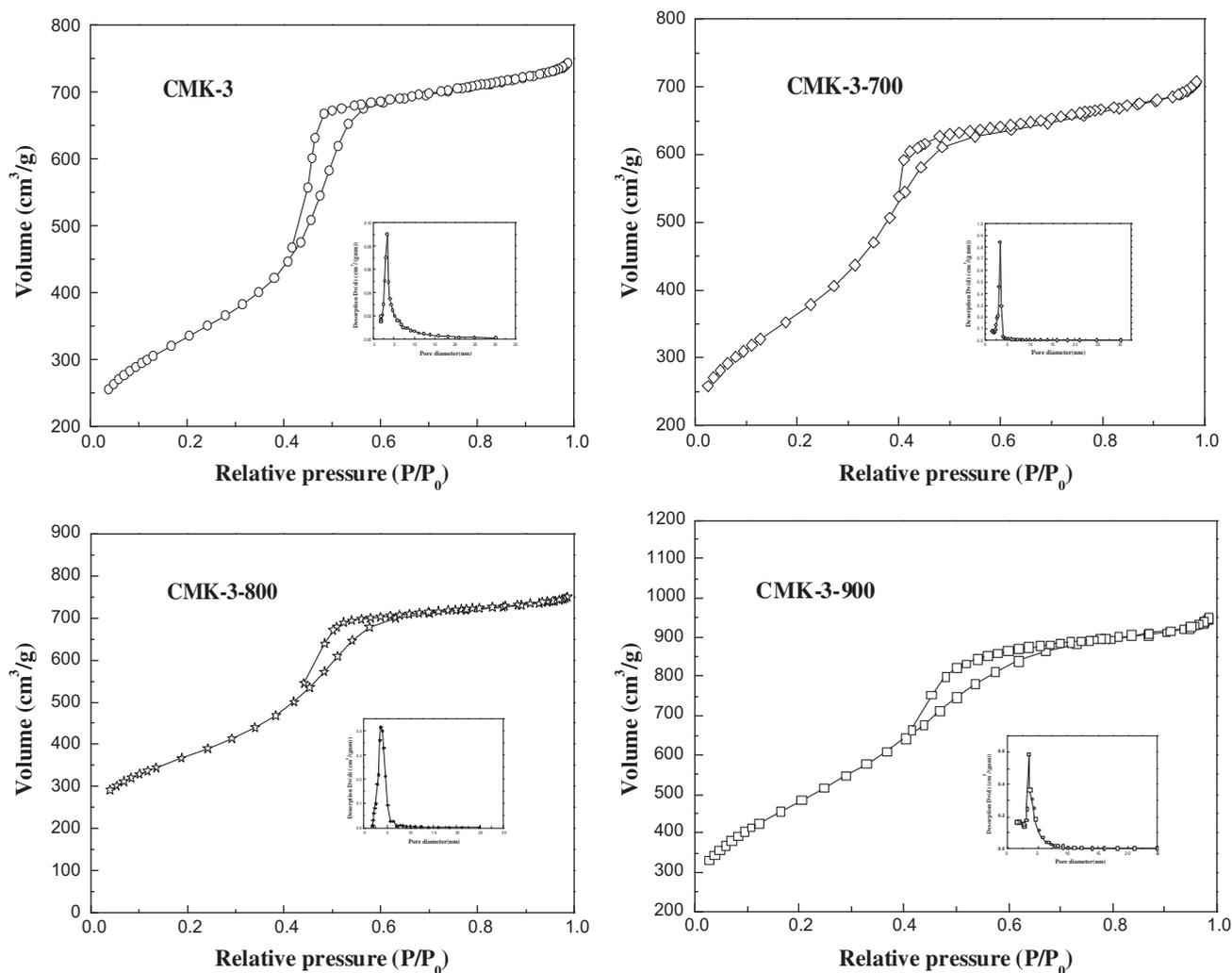


Fig. 2. The  $N_2$  adsorption–desorption isotherm and BJH pore size distribution of original and modified CMK-3.

where they can directly affect adsorbents' adsorption properties. XPS analysis also can further confirm that the major content on the surface is carbon with surface concentration up to 96.37% and there is little nitrogen present on the surface of origin CMK-3 sample. The decreased hydrogen to nitrogen mole ratio of the three modified samples indicated that hydrogen contributes from the oxygen-containing functional groups on the carbon surface which are thermally unstable. Treatment in ammonia causes significant reduction in H/N ratio and this ratio shows a steady decrease upon increasing the severity of ammonia treatment temperature as a result of gradual degradation of the oxygen-containing functional groups. Thus, this phenomenon, as we believe, the enhanced nitrogen content is ascribed to the ammonia modification of nitrogen-containing groups. Hydrogen in the original CMK-3 sample may be mainly from the oxygen-containing

functional groups on the origin CMK-3 surface. Results of XPS studies also indicated that the remaining hydrogen element present in the tailored sample is contributed mainly from the nitrogen functional groups amine and amide with binding energies of 400 and 403.2 eV, respectively [14].

It is also found that the modified reaction temperature plays a key role in the modification process, the amount of nitrogen content increased with an increase in the treatment temperature. The result also indicated that there are more drastic reactions between the radicals from  $NH_3$  decomposition and the CMK-3 surface at higher temperatures. At the same time, the amount of nitrogen-containing functional groups also increased and the amount of the oxygen-containing functional groups decreased. The same trends were observed from XPS analysis results as displayed in Table 3. CMK-3-900 has higher amounts of nitrogen

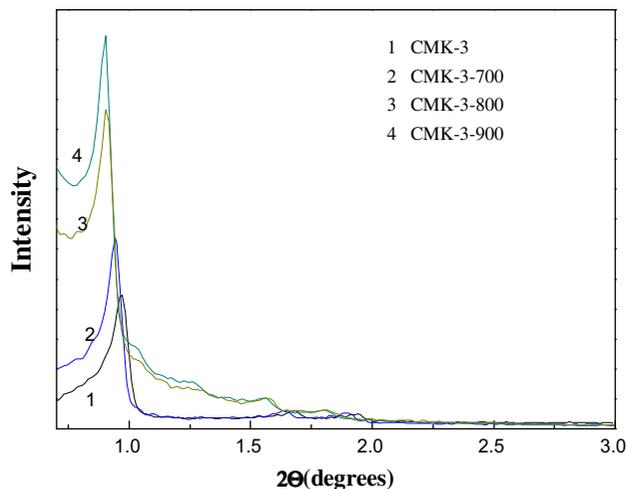


Fig. 3. Low-angle X-ray diffraction patterns of original CMK-3 and modified CMK-3.

content (up to 4.56), as compared to the CMK-3 (0.04), CMK-3-700 (2.81), CMK-3-800 (3.43). But original CMK-3 has higher prime oxygen content than that of

Table 2

Elemental analysis of original CMK-3 and modified CMK-3

Sample	C (wt.%)	N (wt.%)	H (wt.%)	H/N molar ratio
CMK-3	85.34	0.04	1.793	837.85
CMK-3-973	83.45	2.36	1.84	10.92
CMK-3-1073	82.65	3.47	1.67	6.74
CMK-3-1173	82.19	4.58	1.43	4.37

the other modified CMK-3. These oxygen groups will decompose at higher temperatures leaving vacant sites that may be preferentially occupied by ammonia modification. In Table 2, it is observed that a great amount of oxygen-containing functional groups has been removed after modification. There is still a small amount of oxygen functional groups exists on the surface of the treated carbon as the H/N ratio is higher than 4. The observation can be attributed to active sites readsorption of oxygen when samples were washed and exposed to air during drying at 378 K.

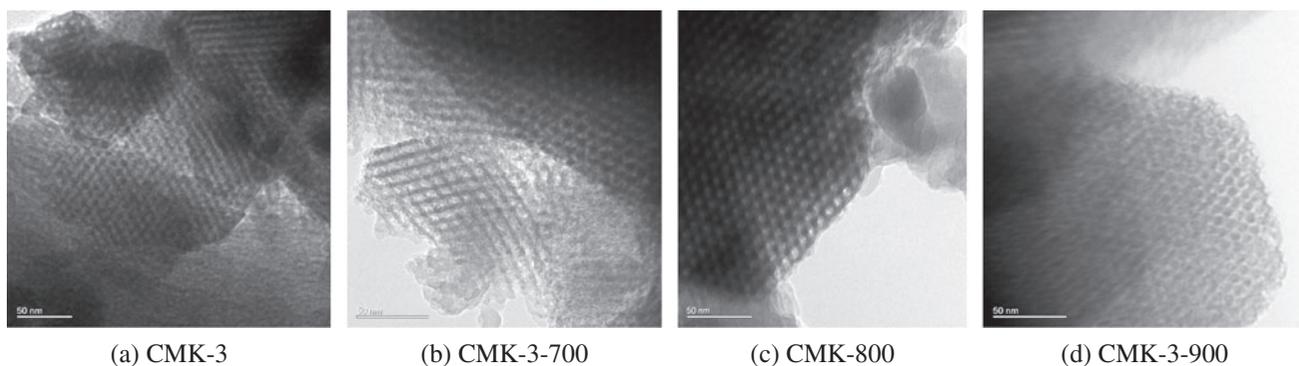


Fig. 4. TEM images of original CMK-3 and modified CMK-3.

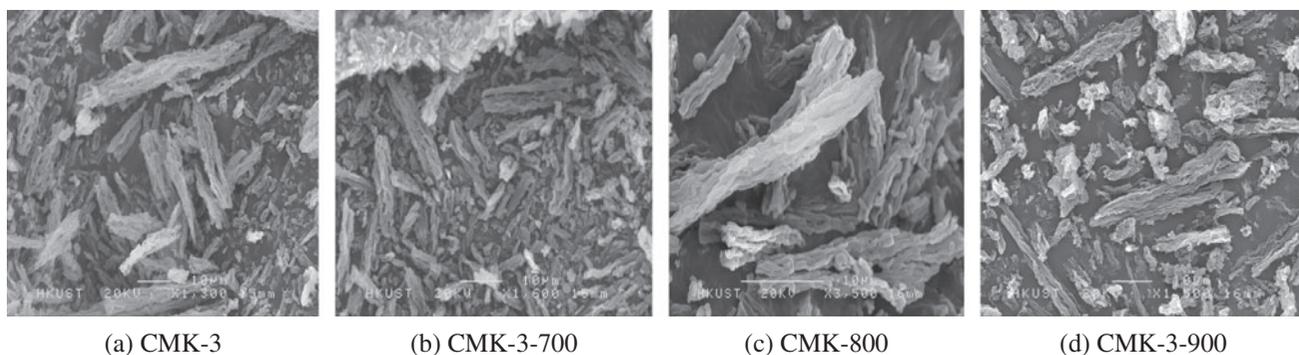


Fig. 5. SEM images of original CMK-3 and modified CMK-3.

Table 3  
XPS of original CMK-3 and modified CMK-3

Sample	C1s (at.%)	N1s (at.%)	O1s (at.%)
CMK-3	96.37	0.04	2.87
CMK-3-973	94.98	2.81	2.02
CMK-3-1073	94.27	3.43	1.73
CMK-3-1173	93.54	4.56	1.55

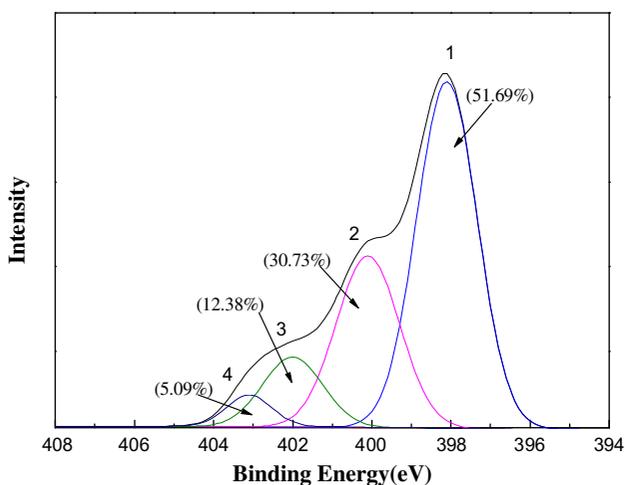


Fig. 6. The representative XPS N1s spectrum for CMK-3-1173 (1) 399.1 eV for pyridine-like nitrogen (2) 400 eV for aromatic amines (3) 402.6 eV for quaternary nitrogen (4) 403.2 eV for protonated amide.

This is in good agreement with the results of XPS analysis (Table 3), indicating that there is still a small amount of oxygen existing on the surface of the modified samples. Similar results have already been reported by Mohammad [18]. As can be seen from Table 4, XPS shows four N1s signals corresponding to nitrogen functional groups formed on modified CMK-3 in the N1s spectral regions, the following binding energies assignments were made: (1) 399.1 eV for pyridine-like nitrogen (2) 400 eV for aromatic amines (3) 402.6 eV for quaternary nitrogen (4) 403.2 eV for protonated amide [19]. Pyridine-like nitrogen and

aromatic amines are the dominant nitrogen-containing functional groups formed on the surface of all the modified samples (four mainly nitrogen-containing functional groups of CMK-3-900 are shown in Fig. 6). This caused for the nitrogen-containing functional groups present at higher temperatures attack the CMK-3, leading to its gasification and the formation of nitrogen functional groups on the surface. Similar XPS result was reported by J.P. Boudou for activated carbon after ammonia modification at high temperatures [20].

### 3.2. Effect of pH

The pH value of solution is one of the most important operational parameters, which can significantly affect the adsorption process especially for dye adsorption [21]. To study the effect of pH on the adsorption process, it is helpful to determine the operational parameters for application. The influence of pH on the adsorption capacity of O II dye onto the original CMK-3 and modified CMK-3 was investigated in the range of pH values from 2 to 12 at an initial dye concentration of 300 mg/L at 25°C. The pH of the solution was adjusted using 0.1 mol/L NaOH or 0.1 mol/L HCl solution.

The effect of pH on the adsorption capacity of O II dye is shown in Fig. 7, it can be observed that the adsorption capacity of the CMK-3 and modified CMK-3 for O II dye decreased as the pH ranges from 2 to 12, the maximum values adsorption of O II dye solution concentrations onto the origin CMK-3 and modified CMK-3 were observed at pH 2.5. Therefore, the optimum pH for the adsorption studies was at pH 2.5. In addition, it was noted that the modified CMK-3 have better adsorption capacity compared with original CMK-3, this may be due to O II dye is one kind of acidic dye, and possesses a strong anionic nature under acidic condition, the increase in the basicity of CMK-3 surfaces will strengthen the interaction between O II dye and modified CMK-3, thus the adsorption capacity of the CMK-3 for O II dye was enhanced [22].

Table 4  
XPS analysis of nitrogen functional groups on original CMK-3 and modified CMK-3 in area %

Sample	Pyridine-like nitrogen (399.1 eV)	Aromatic amines (400 eV)	Quaternary nitrogen (402.6 eV)	Protonated amide (403.2 eV)
CMK-3	–	–	–	–
CMK-3-973	54.57	31.55	10.61	3.26
CMK-3-1073	53.66	30.77	10.55	4.01
CMK-3-1173	51.69	30.73	12.38	5.09

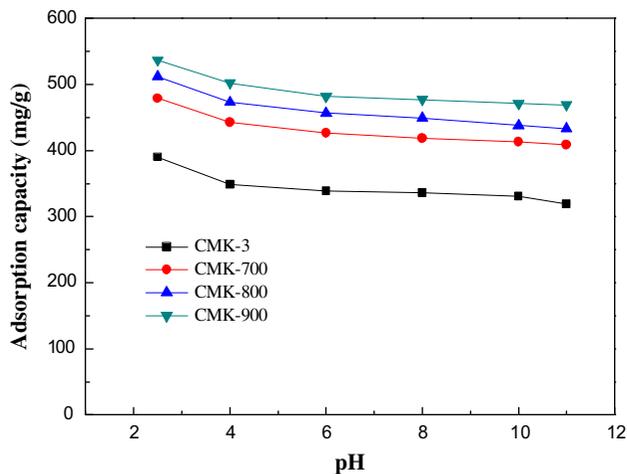


Fig. 7. The effect of pH of dye solution on the adsorption of the O II onto virgin CMK-3 and modified CMK-3 at initial dye concentration of 300 mg/L,  $T = 25^\circ\text{C}$ .

### 3.3. Adsorption studies

The removal efficiency of dye was affected by many physical and chemical factors, such as the type and the structure of dye, the pore texture and surface properties of the adsorbent, and most important the interaction between adsorbent and dye. Some interaction might take place in the adsorption process between the adsorbent and the dye, mainly including, electrostatic interaction, hydrogen bonding, acid–base interaction, and hydrophobic interaction together governing the adsorption process [23]. The presence of basic nitrogen-containing groups has a significant effect on the adsorbents' adsorption properties. In order to better understand the influence of modified CMK-3 for the dye (especially anionic dye) adsorption capacity, the O II dye adsorption test was conducted. Fig. 8 showed adsorption isotherms for the O II dye with original CMK-3 and modified CMK-3.

The equilibrium adsorption capacity in the presence of nitrogen-containing groups making the CMK-3 the better effective adsorbents for O II dye and all of the modified showed a higher adsorption capacity than that of the original CMK-3, and meanwhile the uptake capacity increased as the ammonia modification temperature increases. Fig. 4 shows the original CMK-3 and modified CMK-3 have a very rapid rate in the initial stage of adsorption and gradually decreased with time, and then achieved the adsorption equilibrium in 2.5 h. This was due to the open pore structure of the mesoporous carbons. The adsorption amount of O II dye in the equilibrium can reach 482.18 mg/g for CMK-3-900, which was much higher than that of original CMK-3, CMK-3-700 and

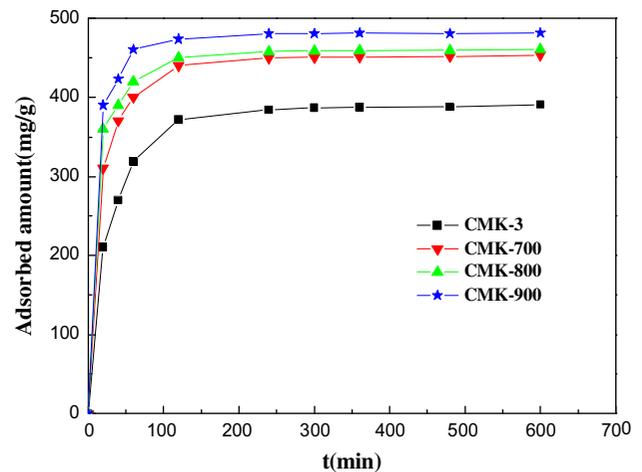


Fig. 8. O II dye adsorption isotherms at  $25^\circ\text{C}$  of original and modified CMK-3.

CMK-3-800. The increasing adsorption amount of O II dye should be related to the change of morphological structure, which is caused by ammonia modification at high temperatures. The comparison between the CMK-3-800 and CMK-3-700 also was investigated. Therefore, the O II dye adsorption capacity of CMK-3 was improved greatly and higher modification temperature was more helpful for O II dye removal.

The presence of nitrogen-containing groups on the surface also may be explained for this phenomenon. These modified CMK-3 (CMK-3-700, CMK-3-800, and CMK-3-900) have higher hydrophobicity than original CMK-3, so O II dye can easily interact with the graphite of modified CMK-3 via hydrophobic interactions [24]. O II dye presents negatively when dissolved in water, so it was expected that O II dye would preferably uptake onto adsorbent with base characteristics. So modified CMK-3 has better adsorption efficiency than that of the original CMK-3, favoring adsorption O II dye (acid dye). The basicity of the adsorbent surface increased as the incorporation of N-containing functional group can strengthen the acid–base interaction between modified CMK-3 and acid dye, favoring the adsorption of cationic dye. The electrostatic interactions are another main adsorption mechanism, there was an enhancement of attractive electrostatic interaction between the anions of O II dye and modified CMK-3 after ammonia modification temperature, as reasons stated above, modified CMK-3-900 has the best adsorption capacity for O II dye [25]. It can be noted that the specific surface area of CMK-3-700 and CMK-3-800 did not greatly improve in comparison with that of CMK-3, but the adsorption capacity increased by 22%. For comparison, the specific surface

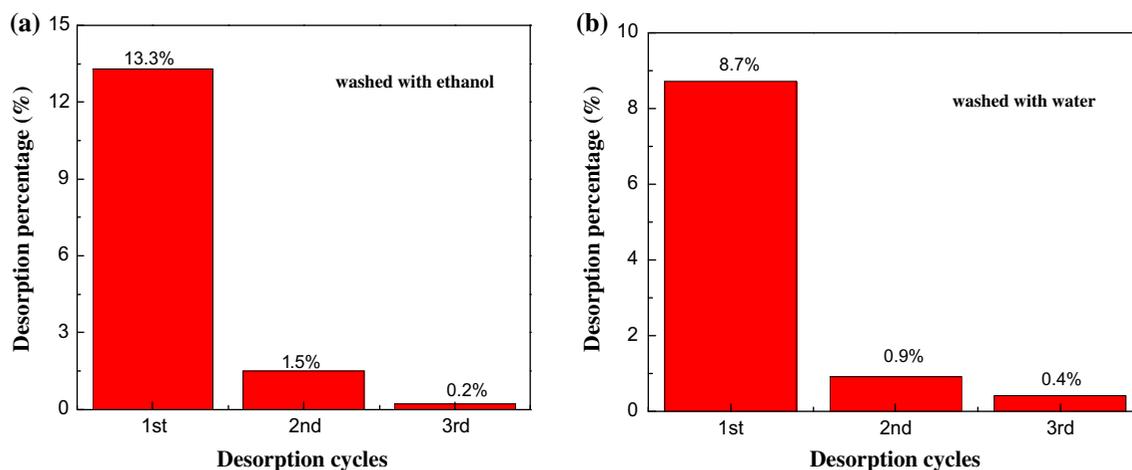


Fig. 9. Desorption of O II dye from modified CMK-3-900 by three desorption cycles ((a) washed with ethanol and (b) washed with distilled water).

area and total volume of CMK-3-700 are the same as those of CMK-3-800, but the adsorption capacity of the latter improved by about 10% compared with the former. Taking into consideration of these observations, the effect of modified CMK-3 in O II dye adsorption may be primarily attributed to N-containing functional groups introduced onto the surface. In conclusion, it could be concluded that the ammonia modification was favorable for enhancing anionic O II dye adsorption capacity.

### 3.3. Desorption studies

Desorption studies is helpful to investigate the stability extent of dyes on the modified CMK-3 and recyclability of spent adsorbent. The recyclability of a dye adsorbent is an important factor. In general, there are two major methods to be used to the recyclability of adsorbent including solvent extraction and thermal annealing at high temperature under inert gas atmosphere (to decompose adsorbed dyes) to regenerate the adsorbent [26]. In this work, the regeneration of the modified CMK-3-900 after dye adsorption was conducted by ethanol extraction and distilled water. The performance of recycled modified CMK-3-900 through three runs of desorption routes is depicted in Fig. 9. It can be observed that O II dye has a higher solubility in ethanol than that in distilled water. The desorption efficiency was low when a small amount of O II dye (around 15 and 10%) could be desorbed from the modified CMK-3-900. Moreover, about 95% of the weakly adsorbed dye was desorbed in the first desorption cycle. The very small amount of O II dye desorbed in the second and third desorption cycles

was negligible. It seemed that adsorption of O II dye on the modified CMK-3 was extremely favorable, tending to be weakly reversible.

## 4. Conclusions

Modified CMK-3 with an orderly uniform pore structure was prepared by a combination of hard templating and ammonia modification. The modified CMK-3 retains the ordered porous structure and has higher surface area, and meanwhile exhibited a better adsorption capacity for O II dye in contrast with original CMK-3. In addition, the desorption test showed that O II dye adsorption on the modified CMK-3 presents good stability.

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