

Research Article

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Adsorption performance of hydrophobic/hydrophilic silica aerogel for low concentration organic pollutant in aqueous solution

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Abstract: Hydrophobic silica aerogels ($\text{SiO}_2(\text{AG})$) was prepared via sol-gel and solvent exchange method under ambient pressure, which could be transformed to hydrophilic after heated under 500°C . Heat treatment cannot change its structure. $\text{SiO}_2(\text{AG})$ samples were the micro-porous structure formed by numerous fine particles and had high specific surface area, pore size and pore volume. The absorption performance of hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ on nitrobenzene, phenol and methylene blue (MB) showed that hydrophobic $\text{SiO}_2(\text{AG})$ exhibited strong adsorption capacity on slightly soluble organic compounds, while hydrophilic $\text{SiO}_2(\text{AG})$ was much more effective on adsorbing soluble compounds, which could be analyzed by the hydrophobic and hydrophilic interaction theory between the adsorbent and adsorbate. Hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ adsorption performance for MB is superior to that for phenol, which could be explained via the electrostatic interaction theory.

Keywords: Silica aerogels; Adsorbent; Organic pollutant

1 Introduction

In the last decades, with the rapid development of industry, large amounts organic compounds were widely used in different field, such as pesticides, dyes, synthetic rubber, plastics. The large-scale use of chemical raw materials has led to a series of environmental pollution. Many or-

ganic pollutants with the feature of bioaccumulation and difficulty biochemical degradation, can present high risk to ecosystem and human health even at low concentration [1, 2]. The prevention and removal organic contaminations in aquatic environment has attracted great attention. Conventional methods like adsorption [3, 4], biodegradation [5], advanced oxidation processes [6–9], etc. have been in-depth investigated and widely used. Compared with other ways, adsorption process is the most commonly technology to remove organic pollutants from water due to its simplicity, high efficiency [3, 4]. In general, the adsorption property of adsorbents is determined by the morphology and structure of porous materials, such as specific surface areas, pore volumes, pore distributions and special pore surface chemistries [10]. Thus, the porous materials with special size and properties, such as graphene, carbon aerogels, activated carbon, polymers, porous silica, and metal-organic frameworks are actively investigated as advanced sorbents [11–16]. Among them, activated carbon is a widely used adsorbent due to its availability and high adsorption capacity [17]. However, carbons display disadvantages such as limited modification flexibility and low selectivity, which limit their applications [18].

As an unique material with wide application [19, 20], silica aerogel ($\text{SiO}_2(\text{AG})$) is a three-dimensional and multi-scaled porous nano-material formed by numerous fine particles and networks. Traditional preparation of $\text{SiO}_2(\text{AG})$ involve two different ways: the process of supercritical drying and ambient pressure drying (APD) technique. The method of supercritical drying can avoid capillary stress and associated drying shrinkage. However, this process is energy intensive and can be dangerous, which leads to high costs and results in limited practical applications [21, 22]. APD technique could overcome the disadvantages of supercritical aerogel process, and made the manufacture and application of $\text{SiO}_2(\text{AG})$ in large scale possible [23–25]. The $\text{SiO}_2(\text{AG})$ has been increasingly researched as an adsorbent owing to its high porosity (up to 99%), high specific surface area, low density, and ease of surface modifi-

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cation, etc. [26–29]. Hrubesh *et al.* [28] prepared hydrophobic $\text{SiO}_2(\text{AG})$ via the supercritical drying method, and proceeded the adsorption experiments of decomposing different organic compounds in water. The results showed that the adsorption capacity of the hydrophobic $\text{SiO}_2(\text{AG})$ was 30 times as that of the carbon for the soluble organic compounds, and up to 130 times for the insoluble organic compounds. Reynolds *et al.* [29] carried out adsorption experiments about hydrophobic $\text{SiO}_2(\text{AG})$ to the oil slick in water, and the results indicated that the capacity of the oil absorption into the hydrophobic $\text{SiO}_2(\text{AG})$ was up to 273 times as its own volume. Perdigoto *et al.* [30] got $\text{SiO}_2(\text{AG})$ from methyltrimethoxysilane (MTMS) as a precursor by supercritical drying, and adsorption capacities of $\text{SiO}_2(\text{AG})$ were shown to uptake more than 50 mg/g of benzene at 50 mg/L of benzene solution. Wang *et al.* [31] prepared hydrophobic $\text{SiO}_2(\text{AG})$ and tested its adsorption for organic compounds from aqueous phase. Adsorption equilibrium of $\text{SiO}_2(\text{AG})$ was reached in under 20 min and the adsorption capacities were 223 mg/g for toluene and 87 mg/g for benzene.

Previous researches suggested that the adsorption capacity of adsorbent was primarily decided by the pore structure and surface area, the surface chemical properties were also important, furthermore, the hydrophobicity, polarity, electron accepting or donating property and the structure of the pollutants could also affect the adsorption affinity [32]. However, the results discussed in many studies on adsorption performance of $\text{SiO}_2(\text{AG})$ has focused on hydrophobic media. Investigation on other factors of adsorption property and adsorption mechanisms were still fragmentary.

In this work, in order to give a fundamental knowledge for potential application of $\text{SiO}_2(\text{AG})$ on pollutants removal. Hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ were prepared via the APD route, and to compare different adsorption performance for nitrobenzene, phenol and methylene blue (MB) with different physicochemical properties, which were common pollutants in industrial wastewater. The adsorption isotherm were evaluated, and the adsorption mechanisms were also discussed.

2 Experimental

2.1 Materials

Tetraethyl orthosilicate (TEOS), anhydrous ethanol (EtOH), hexane (C_6H_{14}), trimethylchlorosilane (TMCS), hydrochloric acid (HCl), ammonia water ($\text{NH}_3 \cdot \text{H}_2\text{O}$), nitrobenzene

($\text{C}_6\text{H}_5\text{NO}_2$), phenol ($\text{C}_6\text{H}_5\text{OH}$) and methylene blue (MB) were analytical grade and commercially purchased from Chron Chemicals Co., Ltd., Chengdu. All reagents were used without any further purification. Deionized water made from laboratory, was used in all experiments.

2.2 Preparation of $\text{SiO}_2(\text{AG})$

Based on the sol-gel method, hydrophobic $\text{SiO}_2(\text{AG})$ was synthesized by controlling the solvent exchanging procedure under ambient pressure. A certain proportion of TEOS, EtOH, and deionized water (mole ratios of TEOS/EtOH/water=1:3:6) were put into a 200 mL beaker, dropping 0.1 mol/L HCl in order to control the pH of the solution in the range of 2.0-3.0. After stirring for 60 min, 0.1 mol/L $\text{NH}_3 \cdot \text{H}_2\text{O}$ solution was added to the silica sol to adjust the pH in the range of 5.0-6.0, until wet gels were gotten. The obtained wet gels were collected and aged for 24 h at room temperature, followed a soaking process in EtOH(50%) for 24 h, then soaked in a mixed solvent with volume ratio of EtOH/TMCS/hexane=1:0.8:1 for 48 h. After filtrating, the wet gels were washed 3 times by hexane and dried 60°C for 24 h, to obtain the hydrophobic $\text{SiO}_2(\text{AG})$. The hydrophilic $\text{SiO}_2(\text{AG})$ was gotten by above processes and calcination at 500°C for 3h [10].

2.3 Characterization

The microstructure of fabricated materials was investigated by SEM (Inspect F, FEI, Holland) and TEM (JEM-2100F, JEOL, Japan). X-ray diffraction (XRD DX-2500) was used to analyzed the crystal structure, the X-ray target made of Cu $K\alpha$ -ray generator (40 kV, 40 mA, $\lambda = 0.15406$ nm). The pore structure and specific surface area (SSA) of the materials were respectively determined by the automatic surface area and porosity analyzer (3H-2000PS4, Beishide Instrument Technology Co., Ltd., Beijing), the test conditions of automatic surface area and porosity analyzer were: nitrogen as adsorbate, degassing mode of heating-vacuum, degassing temperature of 150°C , degassing time of 180 min, saturated steam pressure of 1.0434 bar, ambient temperature of 14.0°C . The surface functional groups of materials were characterized by the FTIR spectra of the materials by using the Fourier transform infrared spectrometer (Nicolet 5700 Spectrophotometer, FTIR), the samples were performed by pressing the power mixed with some of KBr into disks, and the scanning wave number range was set to $4000 \sim 400$ cm^{-1} . The hydrophobic properties of the materials were obtained by measuring contact angle of

water droplets and materials using a contact angle meter (DSA100, KRUSS, Germany).

2.4 Research of adsorption performance

The adsorption experiments were conducted in three different organic solution (nitrobenzene, phenol and methylene blue (MB) solution, respectively). 250 ml conical flask containing 100 ml organic solution and 2 g/l adsorbent (hydrophobic/hydrophilic SiO₂(AG)) were shaken at 220 r/min. Temperature was kept at 25°C. Adsorption capacity of SiO₂(AG) was reflected by measuring the change of concentration of organic compounds, which were determined by UV-vis spectrophotometer (UV-2550, Shimadzu, Japan).

The adsorption amount of organic compounds on adsorbent is calculated as:

$$q = \frac{(c_0 - c_e) \times v}{w} \quad (1)$$

Where q is the adsorption amount of organic compounds on adsorbent, mg/g; C_0 is the initial concentration of organic compounds in solution, mg/l; C_e is the equilibrium adsorption concentration of organic compounds in solution after adsorption equilibrium, mg/l; v is the volume of solution, l; w is the dosage of adsorbent, g.

Langmuir isotherm adsorption model and Freundlich isotherm adsorption model were usually to fit the adsorption behavior of the organic compounds in dilute solution [33].

2.4.1 Langmuir isothermal adsorption model

Langmuir isothermal adsorption model used to describe the adsorption behavior in the solid-liquid system, expressed by equation 2 or 3 [34]:

$$q = \frac{q_m k_L C_e}{1 + k_L C_e} \quad (2)$$

$$\frac{1}{q} = \frac{1}{q_m} + \frac{1}{k_L q_m} \cdot \frac{1}{C_e} \quad (3)$$

Where C_e is the equilibrium concentration in solution, mg/l; q is the adsorption amount in the adsorbent, mg/g; q_m is the saturated adsorption amount in the adsorbent, mg/g; k_L is the Langmuir adsorption constant, l/mg, reflects the affinity between the adsorbate and the adsorption sites.

2.4.2 Freundlich isothermal adsorption model

Freundlich isothermal adsorption model is an empirical equation, which describes the adsorption equilibrium in the solid-liquid adsorption process, that adsorption heat decreased logarithmically with the increases of adsorption amount under isothermal conditions. the model takes uneven surface into account, expressed by equation 4 or 5 [34]:

$$q = K_F C_e^{\frac{1}{n}} \quad (4)$$

$$\ln q = \ln K_F + \frac{1}{n} \ln C_e \quad (5)$$

Where C_e is the equilibrium concentration in solution, mg/l; q is the adsorption amount on the adsorbent, mg/g; K_F is the Freundlich adsorption constant, which is a measure of the adsorption capacity, the greater of its value, then the bigger of the adsorption amount; n is a constant and usually greater than 1, and its inverse is a measure of the adsorption intensity.

3 Results and discussion

3.1 Morphology of SiO₂(AG)

Morphologies of hydrophobic/hydrophilic SiO₂(AG) samples were investigated by SEM and TEM. As shown in Figure 1(a), the hydrophobic SiO₂(AG) samples prepared under ambient pressure presented a spongy-liked shape, which were made of numerous fine particles and formed a loose and porous structure, and uniform particle size distribution. Hydrophilic SiO₂(AG) was gained via hydrophobic SiO₂(AG) calcined 3h under 500°C, Figure 1(b) indicated its surface appearance had no significant difference from hydrophobic SiO₂(AG).

3.2 Specific surface area and pore structure of SiO₂(AG)

The textural properties of SiO₂(AG) were analyzed by using Nitrogen adsorption-desorption method. As can be seen in Figure 2, the N₂ adsorption-desorption isotherms of hydrophobic/hydrophilic SiO₂(AG) samples were a typical type IV adsorption isotherms characteristic with an adsorption hysteresis, indicated that the nano-porous structure exists on hydrophobic/hydrophilic SiO₂(AG), and the hole shape was the narrow tubular pores of the ends open

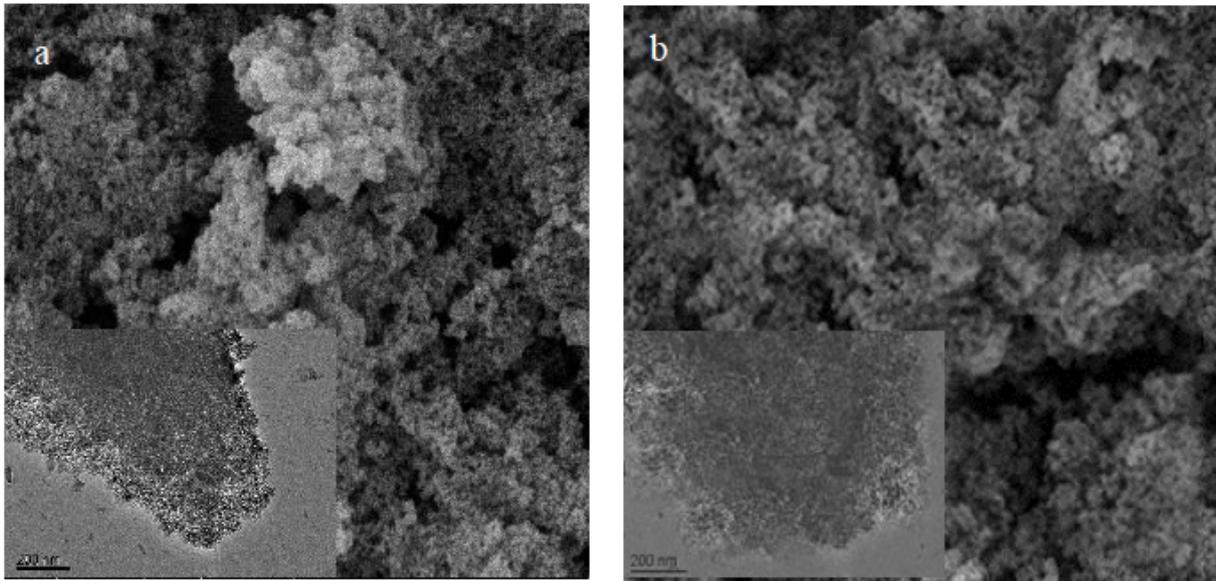


Figure 1: SEM image of SiO₂(AG) sample (a: hydrophobic; b: hydrophilic) (insets are TEM images)

Table 1: SSA, pore size and pore volume of hydrophobic/hydrophilic SiO₂(AG)

Sample	SSA(m ² /g)	pore size (nm)	pore volume (mL/g)
hydrophobic SiO ₂ (AG)	902	8.91	2.0094
hydrophilic SiO ₂ (AG)	928	5.82	1.4085

and mouth width [10]; The SSA was determined by using BET (Brunauer-Emmett-Teller) method. Pore size distribution and total pore volume of the materials were evaluated from the adsorption branch of nitrogen isotherms by using the BJH (Barrett-Joyner-Halenda) method. The insets of Figure 2 displayed the BJH differential integral hole volume and pore size distribution curve of SiO₂(AG).

The experimental results on the SSA, pore size and total pore volume of the SiO₂(AG) powders have been compiled in Table 1. The results showed that hydrophobic/hydrophilic SiO₂(AG) samples had high specific surface area, pore size and pore volume. The conclusions were consistent with the characterization results by TEM. The reason for the decrease in pore size and volume of hydrophilic SiO₂(AG) might be the shrinkage and collapse during the calcination process.

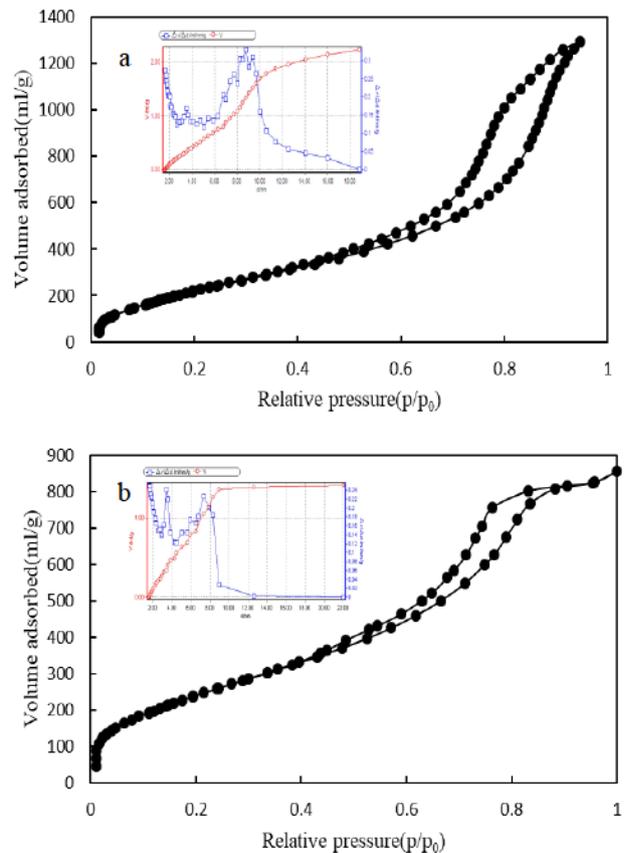


Figure 2: N₂ adsorption-desorption isotherm of SiO₂(AG) (a: hydrophobic; b: hydrophilic) (insets are BJH differential integral hole volume and pore size distribution curve of SiO₂(AG))

3.3 The FT-IR analysis of SiO₂(AG)

The FT-IR spectrum of hydrophobic/hydrophilic SiO₂(AG) were shown in Figure 3. The adsorption peaks at around 3490 cm⁻¹ of hydrophobic SiO₂(AG) was significantly weaker than hydrophilic SiO₂(AG), it was due to that the -OH group on the surface of SiO₂(AG) was replaced by the organic group in the process of modification. As shown in FT-IR spectrum of hydrophobic SiO₂(AG), the characteristic peaks at 2968 cm⁻¹ corresponding to the C-H of -CH₃ antisymmetric stretching vibration, 1387 cm⁻¹ and 928 cm⁻¹ appeared the symmetric vibration and in-plane vibration of -CH₃ group respectively, which indicated -CH₃ to existed on the surface of hydrophobic SiO₂(AG). In addition, the peak at 758 cm⁻¹ corresponding to the symmetric stretching vibration of Si-CH₃, suggested that Si-CH₃ group was contained on the surface of hydrophobic SiO₂(AG). Analyzing by FT-IR, the hydrophobic -CH₃ connected with branched chain of SiO₂(AG) that prepared by solvent exchange method, which let it has hydrophobic properties. The FT-IR of hydrophilic SiO₂(AG) showed the peaks of -OH group existed on the surface of hydrophilic SiO₂(AG), which let it has hydrophilic performance, it was due to that -CH₃ group on the surface of hydrophobic SiO₂(AG) has been translated into -OH group after calcination at 500°C [10].

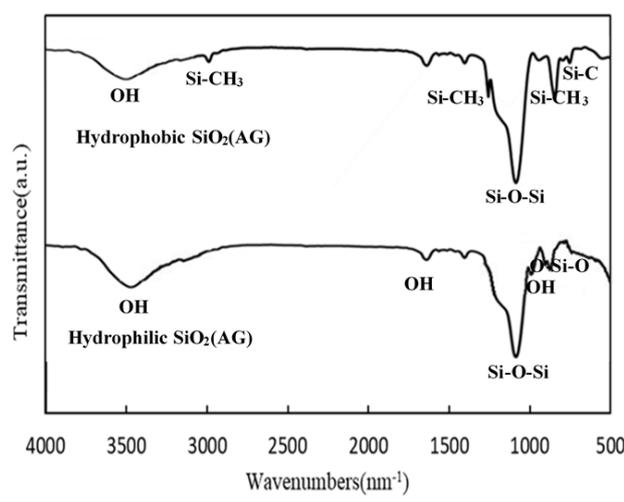


Figure 3: FTIR spectra of hydrophobic/ hydrophilic SiO₂(AG)

3.4 Contact angle of SiO₂(AG)

The hydrophobicity of SiO₂(AG) was measured by testing the contact angle of SiO₂(AG), the results were displayed in

Figure 4. The prepared hydrophobic SiO₂(AG) had a good hydrophobicity, 500°C heat treatment promoted the conversion from hydrophobicity to hydrophilicity. The conclusions were consistent with the characterization results by FT-IR.

3.5 Adsorption property of hydrophobic/hydrophilic SiO₂(AG)

3.5.1 Adsorption curves of different organic compounds

The adsorption experiments were carried out in 100 ml nitrobenzene solution (12 mg/l), phenol solution (10 mg/l) and MB solution (10 mg/l) respectively. The dosage of hydrophobic/hydrophilic SiO₂(AG) was 2.0 g/l. As shown in the adsorption curves of hydrophobic SiO₂(AG) on different adsorbate, Figure 5(a), the hydrophobic SiO₂(AG) exhibited the best adsorption capacity of by removing 51.8% nitrobenzene within 1 h when the system reached adsorption equilibrium, on the other hand, it exhibited poor adsorption capacity on phenol and MB by adsorbing only 9.9% and 17.6%, respectively, and did not get adsorption equilibrium even after 10 h. Figure 5(b) displayed adsorption property of hydrophilic SiO₂(AG), the removal ratio of phenol and MB by adsorption of hydrophilic SiO₂(AG) was 57.8% and 64.3% respectively, and reached adsorption equilibrium within 0.5 h. On the contrary, it showed poor adsorption performance on nitrobenzene by adsorbing only 17.8% and got adsorption equilibrium in 1.5 h.

3.5.2 Adsorption isotherms

Isothermal adsorption experiment were conducted in nitrobenzene solution (12,24,36,48,60 mg/l), phenol solution (10,20,30,40,50 mg/l) and methylene blue (MB) solution (10,20,30,40,50 mg/l), respectively. The liquid adsorption isotherms of hydrophobic/hydrophilic SiO₂(AG) on organic solution, and fitted curves by Langmuir and Freundlich isotherm models were shown in Figure 6(a)-6(c) and Figure 6(d)-6(f), respectively. The results indicated that the equilibrium adsorption amount of hydrophobic/hydrophilic SiO₂(AG) increased with the increase of equilibrium concentration of organic compounds. In the range of the concentration in this experiment, the equilibrium adsorption amount of hydrophobic SiO₂(AG) for nitrobenzene was 6.32 mg/g, while for phenol and MB was 1.19 mg/g and 2.63 mg/g, respectively; but the equilibrium adsorption amount of hydrophilic SiO₂(AG) for nitrobenzene is only 2.38 mg/g, obviously lower than the equilib-

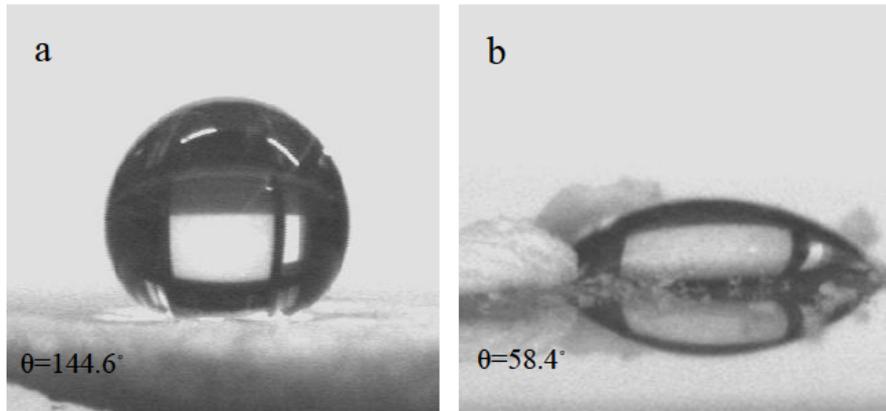


Figure 4: Contact angle of SiO₂(AG) of SiO₂(AG) (a: hydrophobic; b:hydrophilic)

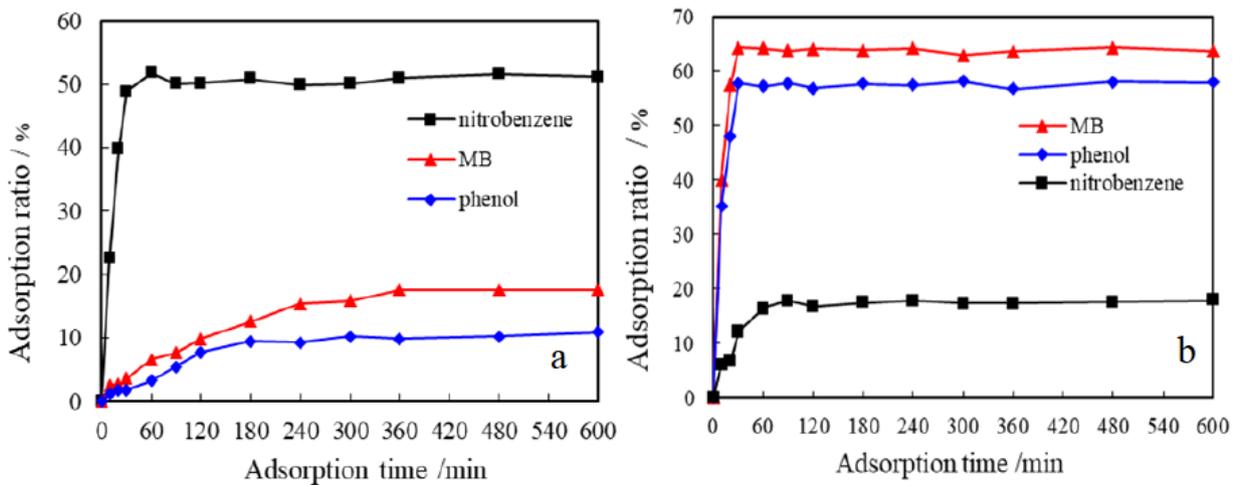


Figure 5: Adsorption curves of nitrobenzene, phenol and MB by SiO₂(AG)

Table 2: Isothermal adsorption fitting results of nitrobenzene, phenol and MB by SiO₂(AG)

adsorbent	adsorbate	Langmuir equation			Freundlich equation		
		q_m	K_L	R^2	K_F	n	R^2
hydrophobic SiO ₂ (AG)	nitrobenzene	6.9686	0.1415	0.9866	1.8621	3.1221	0.9904
	phenol	1.7322	0.0454	0.9958	0.1631	1.8975	0.9969
	MB	4.6468	0.0286	0.9976	0.2436	1.5816	0.9833
hydrophilic SiO ₂ (AG)	nitrobenzene	3.0506	0.0557	0.9862	0.3953	2.2148	0.9927
	phenol	6.9348	0.1718	0.9591	1.7736	2.7601	0.9973
	MB	9.5329	0.1449	0.9954	1.9883	2.3981	0.9910

rium adsorption amount of hydrophobic SiO₂(AG) for nitrobenzene, while for phenol and MB reached 6.40 mg/g and 8.32 mg/g respectively, significantly higher than the equilibrium adsorption amount of hydrophobic SiO₂(AG).

The relevant parameters of Langmuir and Freundlich isotherm models were displayed in Table 2. The results

showed that Langmuir and Freundlich isotherm models could fit well the adsorption isotherms of SiO₂(AG) for nitrobenzene, phenol and methylene blue. Related studies [34] suggested that Freundlich constant K_F was a constant related to the adsorption capacity, the greater K_F would course greater adsorption; when the value of con-

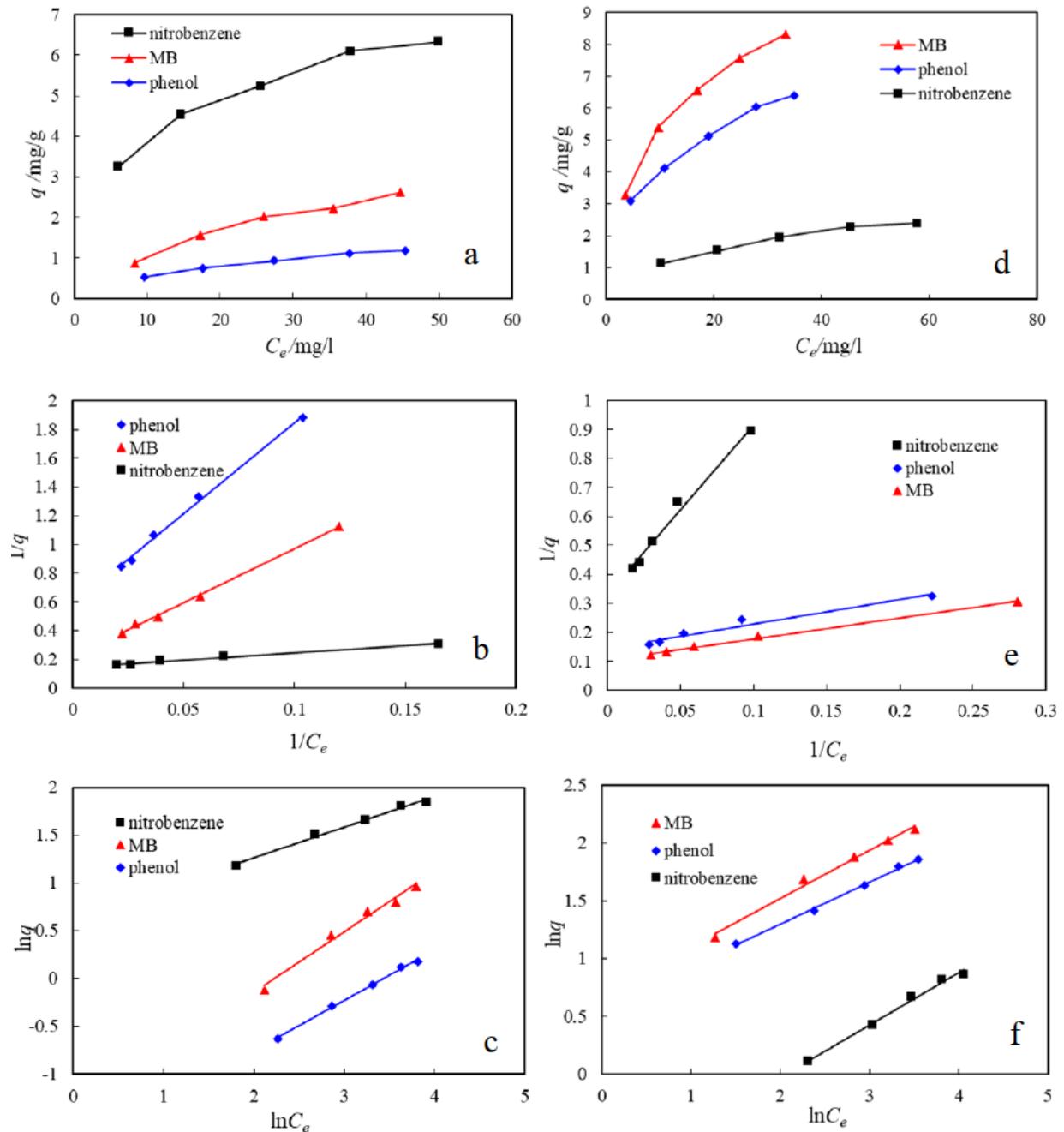


Figure 6: Adsorption isotherm and fitting curves of nitrobenzene, phenol and MB by SiO₂(AG) (a), (b), (c) and (d), (e), (f): Adsorption isotherm, Langmuir fitting curves and Freundlich fitting curves of hydrophobic SiO₂(AG) and hydrophilic SiO₂(AG), respectively

stant $1/n$ was in the range from 0.1 to 0.5, the adsorbate was easily absorbed, and the value was smaller, the adsorption properties were better; with the increases of $1/n$, while the equilibrium concentration of the adsorbate was higher, the absorption capacity would play more sufficient. Compared with the relevant parameters from Table 2, adsorption performance of hydrophobic SiO₂(AG) for nitrobenzene was much better than that for phenol and MB; while

the hydrophilic SiO₂(AG) expressed a better adsorption property for phenol and MB; Both the hydrophobic and hydrophilic SiO₂(AG), its adsorption properties for MB were superior to that for phenol.

3.5.3 Adsorption mechanism research

In the organic compounds adsorption process in solution, the adsorbent surface area, pore size distribution, pore structure and other characteristics of material plays a decisive role to its adsorption properties [35]. In addition, adsorbents exhibit differences of adsorption performance to different adsorbates, which could be analyzed by the hydrophobic and hydrophilic interaction theory between the adsorbent and adsorbate, molecular polarity theory, electrostatic interaction theory, the electron donor-acceptor interaction theory, π - π dispersion theory [36, 37]. According to the hydrophobic and hydrophilic interaction theory in water phase, the hydrophobicity of $\text{SiO}_2(\text{AG})$ plays a leading role in the organic compounds adsorption process. Nitrobenzene is an insoluble hydrophobic organic compounds, MB and phenol are hydrophilic organic compounds and freely soluble in water, and the hydrophilic $\text{SiO}_2(\text{AG})$ is infiltrated easily by water molecules which occupy a part of pores. So, the hydrophobic $\text{SiO}_2(\text{AG})$ shows better adsorption to nitrobenzene than other one; hydrophilic $\text{SiO}_2(\text{AG})$ has better adsorption to MB and phenol. Hydrophobic and hydrophilic $\text{SiO}_2(\text{AG})$ adsorption performance for MB is superior to that for phenol, basing on the electrostatic interaction theory, SiO_2 isoelectric point is very low [38], less than the pH of the phenol and MB solution, $\text{SiO}_2(\text{AG})$ in phenol and MB solution will show electro-negativity, MB is a typical cationic dye with a positive charge, while phenol is electrically neutral. So there exist an electrostatic attraction between MB and $\text{SiO}_2(\text{AG})$, and the adsorption performance of $\text{SiO}_2(\text{AG})$ for MB prove better than phenol.

4 Conclusion

Via sol-gel and solvent exchange method, hydrophobic $\text{SiO}_2(\text{AG})$ was prepared under ambient pressure, and it could be transformed to hydrophilic after 500°C heat treatment. The results of material characterization showed that the synthesized hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ were the micro-porous structure formed by numerous fine particles and had high specific surface area, pore size and pore volume.

The adsorption experiments of hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ for nitrobenzene, phenol and MB showed that hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ was effective in removing organic compounds from aqueous solution. Hydrophobic $\text{SiO}_2(\text{AG})$ showed much stronger adsorption effectiveness on slightly soluble organic com-

pounds, while hydrophilic $\text{SiO}_2(\text{AG})$ was much more effective on adsorbing soluble compounds, which could be analyzed by the hydrophobic and hydrophilic interaction theory between the adsorbent and adsorbate. Adsorption performance of hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ for MB is superior to that for phenol, which could be explained via the electrostatic interaction theory.

Due to the high adsorption efficiency on a wide range of organic pollutant in water, easy modification of the properties, and the relative low manufacture cost, hydrophobic/hydrophilic $\text{SiO}_2(\text{AG})$ has great application potential in the wastewater treatment field.

Data accessibility: We have conducted our experiment systematically and reported their experimental procedure clearly in the experimental section and provided all necessary data in results and discussion section in the main manuscript.

Author Contributions: Zhigang Yi designed, supervised experiments and led the drafting of manuscript. Qiong Tang and Tao Jiang assisted in the analysis and testing work during the experiments. Ying Cheng contributed to characterization of property of material.

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