Preparation and Properties of Highly Transparent SiO\(_2\) Aerogels for Thermal Insulation

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Abstract: SiO\(_2\) aerogels have attracted extensive attention due to their unique structural characteristics, which exhibit many special properties, especially good optical transparency. As far as we know, the sol-gel stage during the synthesis of aerogel plays an important role in the construction of the gel skeleton. In this study, we adjusted the amount of silicon source and catalyst to explore the best scheme for preparing highly transparent SiO\(_2\) aerogels, and further clarify the effects of both on the properties of SiO\(_2\) aerogels. Results indicated that the pore size distribution was between 10 and 20 nm, the thermal conductivity was between 0.0135 and 0.021 W/(m·K), and the transmittance reached 97.78% at 800 nm of the aerogels, better than most studies. Therefore, it has the potential to be used in aerogel glass for thermal insulation.

Keywords: SiO\(_2\) aerogels; high transparency; thermal insulation; mesoporous materials

1. Introduction

Aerogel [1] is a typical thermal insulation material with a three-dimensional (3D) porous network structure connected by nanoparticles. It has been widely used in numerous fields due to its many unique characteristics and properties, such as low density, high porosity, extremely low thermal conductivity and excellent acoustic properties. SiO\(_2\) aerogel [2] with excellent optical transparency has emerged as the most representative aerogel due to its inexpensive cost and straightforward preparation process [3–6]. Transparent SiO\(_2\) aerogel [7,8] has been utilized extensively in Cherenkov detectors [9,10], thermal insulation [11–15], energy-saving [13,16–19], photoelectric materials [20–22], wearables fields [23], thermal collectors [24–28], adsorbents and sensors. Thus, transparent SiO\(_2\) aerogel has great research value and the prospects are very broad.

The thermal insulation performance of aerogels has been demonstrated to be due to the low thermal conductivity provided by the high porosity structure [29] and the pore size is preferably smaller than the air free path (70 nm). In addition, high transparency performance requires that the pores of aerogel be much smaller than the wavelength of visible light, so as to minimize the scattering of block. As a result, the porous size of aerogels with thermal insulation as well as highly transparent must be small and uniformly distributed to avoid large deviation of light at the interface of pores. Moreover, SiO\(_2\) aerogels are generally prepared by the sol-gel method. Due to the internal disordered porous structure, they are easily cracked and broken during the preparation process, and it is a big challenge to obtain the overall block structure without defects [30]. In most research, the supercritical drying method without gas-liquid interface is used to avoid the generation of capillary forces and reduce the shrinkage of gel.

Researchers have been trying to improve the optical transparency of SiO\(_2\) aerogels, but it is exceedingly difficult to combine thermal insulation with high transparency [31–34].
At present, there are many mainstream or mature strategies to improve the transmittance of SiO$_2$ aerogels. Firstly, in the sol-gel stage, Shimizu et al. [35] found, based on surfactants, that different concentrations of the base catalyst affected the size and uniformity of aerogels skeleton, and thus affected transparency. Zhao et al. [24] proposed a rapid-hydrolysis-condensation procedure and restricted-cluster, and the molar ratio of catalyst was identified as the most critical parameter in the experimental process in order to obtain highly transparent aerogels. Moreover, during the drying process after sol-gel, by controlling shrinkage in the aging process of the gels, Lei et al. [36] prepared polyesiciloxane aerogels with smaller particles and pores. The aerogels with the density of 0.25 g/cm$^3$ had a transmittance of 70% at 550 nm. Similarly, Tabata et al. [9] prepared SiO$_2$ aerogels with higher transparency than conventional methods using a method called pinhole drying [37]. Despite the fact that the transmittance was improved and there were no cracks in the appearance of aerogels, the shrinkage rate of aerogels after drying was very high, which was a common problem in atmospheric drying [1,21]. Finally, after the preparation stage, high-temperature annealing is one of the effective methods to change the microstructure of aerogels. Strobach et al. [25,26] used the viscosity change of aerogels at high temperature to continue to improve transparency. The results showed that aerogels were relatively stable at a low temperature, but particles tend to be extremely aggregated and densified at high temperature. Hence, the size of the scattering source also had a great dependence on temperature, resulting in the increase of transparency. Although temperature treatment can significantly improve transparency, high-temperature treatment is time-consuming and laborious, and will cause shrinkage of aerogels, resulting in an increase in thermal conductivity. In general, the monolithic SiO$_2$ aerogel system became a promising material for windows with highly energy-efficient [12,14,38].

In this paper, we aim to prepare highly transparent SiO$_2$ aerogels with a concise and efficient method. Thus, the alkaline one-step sol-gel method was adopted, and tetramethoxysilane (TMOS), a silicon source with a small molecular volume was selected, and by adjusting the amount of silicon source and alkali catalyst to simply regulated the gel structure. Lastly, the highly transparent SiO$_2$ aerogels were obtained by the appropriate drying method. Here, no additional surfactants and post-treatment were required, the SiO$_2$ aerogels with great advantages in optical transparency were directly obtained through simple processes. Compared to glass, SiO$_2$ aerogels do not generate glare spots, and hold great promise as a means of thermal insulation in the interspace of double-glazing systems as well as in solar collectors.

2. Results and Discussion

2.1. FTIR Spectrum Analysis

Figure 1 showed the FTIR spectrum of SiO$_2$ aerogels, in which 3422 cm$^{-1}$ was the antisymmetric stretching vibration peak of -OH and 1627 cm$^{-1}$ was the bending vibration peak of H-O-H; 1094 cm$^{-1}$ was the antisymmetric stretching vibration peak of the Si-O-Si group, which belonged to the characteristic peak of SiO$_2$, indicating that the main component of sample was SiO$_2$; 957 cm$^{-1}$ was the stretching vibration peak of Si-OH, and this group was the main reason for the hydrophilicity of SiO$_2$ aerogel; 802 cm$^{-1}$ was the symmetric stretching vibration peak of Si-O, which also belonged to the characteristic peak of SiO$_2$.

2.2. Microstructure Analysis

The micromorphology of SiO$_2$ aerogels was a typical nano-porous 3D network structure. As shown in Figure 2, when the concentration of NH$_4$OH solution was 0.75 mol/L and the mass fraction of TMOS was 15 wt.%; 25 wt.%; 35 wt.% and 45 wt.%, the micromorphologies with continuous porosity of the prepared SiO$_2$ aerogels were very similar. Small and uniform particles were closely connected with each other through the "neck", and the structure was relatively dense. Due to the different mass fractions of TMOS, the aerogels occurred different shrinkage after drying, as shown in Table 1, resulting in a
secondary change in the microstructure. With the decrease in the mass fraction of silicon source, the shrinkage gradually increased, and the structures tended to be dense, which made the structure of SiO$_2$ aerogels with high and low mass fraction very similar, and still maintained a pearl-like 3D network structure. The increase in TMOS content only deepened the densification of the structure slightly. Therefore, the SiO$_2$ aerogels prepared under the condition of different TMOS mass fractions presented a small difference in the micromorphology.

![FTIR spectrum of SiO$_2$ aerogels.](image)

**Figure 1.** FTIR spectrum of SiO$_2$ aerogels.

**Table 1.** Shrinkage of SiO$_2$ aerogels with different mass fractions of TMOS.

| TMOS (wt.%) | 15   | 25   | 35   | 45   |
|-------------|------|------|------|------|
| Line Shrinkage (%) | 22–27| 11–15| 8–10 | 5–9  |

However, when the mass fraction of TMOS was 45 wt.%, and the concentration of NH$_4$OH solution was 0.15 mol/L; 0.30 mol/L; 0.45 mol/L; 0.60 mol/L and 0.75 mol/L, the size of SiO$_2$ particles and pores decreased with the increase in the concentration of NH$_4$OH solution, and the structure tended to be denser, as shown in Figure 3. It was attributed to the high catalyst concentration improving the speed of gelation, so that the silicon source can rapidly hydrolyze and condense into primary particles. Then, due to the depletion of the silicon source, the formation of secondary particles lacked raw materials at the later stage, so the clustering process stopped abruptly, resulting in smaller SiO$_2$ particles, smaller pores, and more uniform distribution [24].
Moreover, as shown in Figure 4, the effect of silicon source content and catalyst concentration on the microstructure was also illustrated. Figure 4a showed the N\textsubscript{2} adsorption-desorption isotherm curves of SiO\textsubscript{2} aerogels which belonged to the type IV, a typical characteristic of mesoporous materials [21]. With the increase in TMOS mass fraction, the specific surface area gradually decreased. On the contrary, with the increase in NH\textsubscript{4}OH concentraton on the microstructure was also illustrated. Figure 4a showed the N\textsubscript{2} adsorption-desorption isotherm curves of SiO\textsubscript{2} aerogels which belonged to the type IV, a typical characteristic of mesoporous materials [21]. With the increase in TMOS mass fraction, the specific surface area gradually decreased. On the contrary, with the increase in NH\textsubscript{4}OH.

Figure 2. SEM of SiO\textsubscript{2} aerogels: the concentration of NH\textsubscript{4}OH solution was 0.75 mol/L and the mass fractions of TMOS were (a) 15 wt.%; (b) 25 wt.%; (c) 35 wt.%; (d) 45 wt.%. Moreover, as shown in Figure 4, the effect of silicon source content and catalyst concentration on the microstructure was also illustrated. Figure 4a showed the N\textsubscript{2} adsorption-desorption isotherm curves of SiO\textsubscript{2} aerogels which belonged to the type IV, a typical characteristic of mesoporous materials [21]. With the increase in TMOS mass fraction, the specific surface area gradually decreased. On the contrary, with the increase in NH\textsubscript{4}OH.

Figure 3. SEM of SiO\textsubscript{2} aerogels: the mass fraction of TMOS was 45 wt.% and the concentration of NH\textsubscript{4}OH solutions were (a) 0.15 mol/L; (b) 0.30 mol/L; (c) 0.45 mol/L; (d) 0.60 mol/L; (e) 0.75 mol/L. Moreover, as shown in Figure 4, the effect of silicon source content and catalyst concentration on the microstructure was also illustrated. Figure 4a showed the N\textsubscript{2} adsorption-desorption isotherm curves of SiO\textsubscript{2} aerogels which belonged to the type IV, a typical characteristic of mesoporous materials [21]. With the increase in TMOS mass fraction, the specific surface area gradually decreased. On the contrary, with the increase in NH\textsubscript{4}OH.
solution concentration, the specific surface area increased accordingly. This was mainly because more silicon sources, the hydrolysis and condensation reaction will be more sufficient, and more SiO<sub>2</sub> particles can be generated to secondary particles, as well as the catalyst enabling the reduction of SiO<sub>2</sub> particle size by rapid gel process. As shown in Figure 4b, it can be seen that the pore size distribution of each sample was mainly concentrated between 10 and 20 nm, which was divided obviously into two concentrated points due to the different concentration of NH<sub>4</sub>OH solution. When the concentration of NH<sub>4</sub>OH solution was low, it was between 15 and 20 nm with wide distribution, and when the concentration of NH<sub>4</sub>OH solution was high, it is between 10 and 15 nm with narrow distribution. However, the pore size of the NH<sub>4</sub>OH solution with the same concentration and different TMOS content was roughly the same. Therefore, the effect of NH<sub>4</sub>OH solution concentration on the pore size distribution was more significant.

2.3. Thermal Properties

As shown in Figure 5a, there are three ways of heat conduction in aerogels: solid-phase heat transfer, gas convection heat transfer, and radiation heat transfer. In general, radiation heat transfer is ignored at room temperature [1]. Figure 5b showed the thermal conductivity of SiO<sub>2</sub> aerogels. It can be seen that with the increase in TMOS mass fraction, the thermal conductivity increased, because higher TMOS mass fraction led to higher bulk density, resulting in an increase in the solid-phase heat conduction of the network skeleton. However, the thermal conductivity of 15 wt.% aerogels was slightly higher than that of 25 wt.%. Maybe because too little TMOS content led to larger pores, resulting in higher gas convection heat transfer. At the same time, when the mass fraction of TMOS was same, the thermal conductivity decreased with the increase in the concentration of NH<sub>4</sub>OH solution. This was owing to a high catalyst concentration, which led to smaller SiO<sub>2</sub> particles and pores, which limited the solid-phase heat transfer and gas convection heat transfer, resulting in a lower final thermal conductivity.
The thermal conductivities of SiO\(_2\) highest thermal conductivity and had a great advantage in thermal insulation.

Figure 6. The infrared thermal imaging of the SiO\(_2\) aerogel. It can be seen that the surface temperature of the SiO\(_2\) aerogel tended to be stable after 300 s, and the temperature difference \(\Delta T = 44^\circ\text{C}\) at 600 s, which was about 40% of panel temperature, indicating that the highly transparent SiO\(_2\) aerogel had excellent thermal insulation performance.

Table 2 listed the thermal conductivities of SiO\(_2\) aerogels, xerogel and cryogel. Through comparison, we found that thanks to the supercritical drying method, SiO\(_2\) aerogel had the lowest thermal conductivity and had a great advantage in thermal insulation.

Table 2. The thermal conductivities of SiO\(_2\) aerogels, xerogel and cryogel [29,30,39,40].

| Samples | SiO\(_2\) Aerogel | SiO\(_2\) Xerogel | SiO\(_2\) Cryogel |
|---------|------------------|------------------|------------------|
| Thermal conductivity (W/(m·K)) | 0.01–0.03 | 0.03–0.09 | 0.03–0.05 |
2.4. Optical Transmittance

According to the Rayleigh-Gans theory, the extinction coefficient of aerogels is proportional to the bulk density of aerogels and the diameter of the scattering source [7], as follows:

\[
\sigma_s = 4\pi^4 \frac{\rho_{ap}}{\rho_{SiO_2}} \frac{d^3}{\lambda^4} \left( \frac{n_1^2 - 1}{n_2} \right)
\]

where \( \sigma_s \) is scattering coefficient, \( \rho_{ap} \) is apparent density or bulk density of SiO\(_2\) aerogels, \( \rho_{SiO_2} \) is true density of amorphous silicon (2.2 g/cm\(^3\)), \( d \) is diameter of particle scattering center, \( \lambda \) is wavelength, \( n \) is relative refractive index (\( n_1 \) is refractive index of the SiO\(_2\) particles, \( n_2 \) is refractive index of the external medium (air)). The formulation applies to isolated SiO\(_2\) spheres, which approximate a highly porous bulk.

In detail, one of the scattering sources is the macroscopic defects of appearance, which may be caused during the sol-gel process or demolding process [16], so the whole preparation process should try to ensure no pollution and destruction. Another scattering source is the scattering of nano-porous structures, which is the result of the interaction between SiO\(_2\) particles and pores [16]. In fact, we can control the density of SiO\(_2\) aerogels and the diameter of the scattering source, but because the relationship between the diameter of the scattering source and the extinction coefficient is a power of 3, it becomes a more critical factor.

As shown in Figure 7, SiO\(_2\) aerogels themselves have a very low absorption of light. When SiO\(_2\) aerogels are irradiated by a beam of light, it may interact with the scattering source in the aerogels and then weaken the intensity of transmittance. According to the Rayleigh-Gans theory, the smaller the size of the SiO\(_2\) aerogels structural unit, the smaller the extinction coefficient and the higher the transmittance [41]. Therefore, the optical transparency of SiO\(_2\) aerogels was related to the gel structure construction [41,42]. The optimization of composition and microstructure control are effective strategies to improve the high performance and multi-performance of SiO\(_2\) aerogels [1].

![Diagram of light transmittance](image)

**Figure 7.** The propagation paths of light in SiO\(_2\) aerogels.

Figure 8 showed that the transmittance curves of SiO\(_2\) aerogels. Figure 8a–d were the transmittance curves under the same TMOS mass fraction and different NH\(_4\)OH solution concentrations, respectively. According to Figure 8c–f, the results showed that the transmittance of SiO\(_2\) aerogels prepared by 45 wt.% TMOS and 0.75 mol/L NH\(_4\)OH solution were the highest, which reached 97.78% at 800 nm. The reason for that was because the high content of silicon source led to sufficient SiO\(_2\) particles generated, while the high concentration of catalyst promoted the hydrolysis and condensation reaction of SiO\(_2\), and the cluster was restricted. Under these conditions, the generated SiO\(_2\) particles and pores were small and uniform, the scattering source was small, which reduced the deviation of...
light as well as the scattering of light in the aerogels, so the transparency of aerogels was the highest. The results were consistent with the previous analysis.

Figure 8. Transmittance curves of SiO$_2$ aerogels with different TMOS mass fractions: (a) 15 wt.%; (b) 25 wt.%; (c) 35 wt.%; (d) 45 wt.%; (e) the highest transmittance among the TMOS mass fractions of 15–45 wt.% and its (f) local magnification of wavelengths from 750 nm to 850 nm.
3. Conclusions

We used different mass fractions of TMOS and different concentrations of NH$_4$OH solution to simply control the structure of SiO$_2$ aerogels, thereby improving the transmittance. The micromorphology including densification, particle size and pore size distribution of SiO$_2$ aerogels were closely related to the silicon source content and catalyst concentration. After adjusting the experimental parameters, the particles and pore size distribution of SiO$_2$ aerogels were small, and the thermal conductivity was low. More notably, the highest transmittance of SiO$_2$ aerogels can reach 97.78% at 800 nm, which was higher than that of ordinary SiO$_2$ aerogels and undoubtedly broaden the application prospect of SiO$_2$ aerogels in highly transparent materials, such as aerogel glass. However, the mechanical properties and hydrophilicity of highly transparent SiO$_2$ aerogels should be further modified for improvement.

4. Materials and Methods

4.1. Materials

TMOS (AR) was purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. (Shanghai, China). Methanol (AR) was purchased from Beijing Tongguang Fine Chemical Company (Beijing, China). The water (deionized water) was purchased from Shanghai Yien Chemical Technology Co., Ltd. (Shanghai, China). NH$_4$OH (GR) was purchased from Shanghai Maclin Biochemical Technology Co., Ltd. (Shanghai, China). Ethanol (AR) was purchased from Beijing Tongguang Fine Chemical Company (Beijing, China). All chemical regents were used without further purification.

4.2. Preparation of SiO$_2$ Aerogels

The schematic of the preparation process was shown in Figure 9 and the reaction process was shown in Figure 10. The preparation formulations of SiO$_2$ aerogels were listed in Table 3. In the experiments, the mass ratios of TMOS were 15, 25, 35 and 45 wt.%, and the concentrations of NH$_4$OH solution were 0.15, 0.30, 0.45, 0.60 and 0.75 mol/L. Firstly, a certain amount of TMOS and methanol were mixed at room temperature and stirred for 30 min. Then, NH$_4$OH solutions were added and continued to stir until uniform and moved to the mold, which needed to be sealed and placed in a 30 $^\circ$C oven for 24 h. After the demolding process, the gel was soaked in ethanol for 3 days for solvent replacement and cleaning. Finally, supercritical CO$_2$ drying [43] was performed to obtain highly transparent SiO$_2$ aerogels. The specific parameters of supercritical CO$_2$ drying were set as follows: the temperature of the drying kettle was 32 $^\circ$C, the temperature of the separating kettle was 37 $^\circ$C, the pressure of the storage tank was 5.5 MPa, the drying pressure was 7.5 MPa, the separation pressure was 5.5 MPa, and the supercritical state was maintained for about 1 h. Parameters can be adjusted according to different aerogel formulations. Figure 11 showed the pictures of the prepared SiO$_2$ aerogels with different transparency.

4.3. Characterizations

The microstructure of SiO$_2$ aerogels was observed using a Hitachi S4800 scanning electron microscope. Nitrogen adsorption-desorption tests for observation of pore structure were performed using a Micrometeritics APSP 2460 automatic specific surface area and porosity analyzer. The DRPL-III high-precision thermal conductivity tester was used to test the thermal conductivity of the SiO$_2$ aerogels; the temperature of the hot surface was 40 $^\circ$C, the temperature of the cold surface was 20 $^\circ$C, and the pressure was 5 N. A TU-1901 double-beam UV-Vis spectrophotometer was used to perform spectral scanning on the SiO$_2$ aerogels to obtain the transmittance (T%), and the test wavelength range was 200–900 nm. The SiO$_2$ aerogels were formed into a size of 80×80 mm and the thickness was 2–3 mm.
Figure 9. Schematic of preparation process of highly transparent SiO$_2$ aerogels.

(a) Hydrolysis reaction of TMOS:

\[ \text{H}_3\text{CO-Si-OCH}_3 + 4\text{H}_2\text{O} \rightarrow \text{HO-Si-OH} + 4\text{CH}_3\text{OH} \]

(b) Condensation reaction of TMOS after hydrolysis:

\[ 2\text{HO-Si-OH} \rightarrow \text{HO-Si-O-Si-OH} + \text{H}_2\text{O} \]

(c) Further cross-linking by condensation reaction to form a 3D network.

Figure 10. (a) Hydrolysis reaction of TMOS; (b) condensation reaction of TMOS after hydrolysis; (c) further cross-linking by condensation reaction to form a 3D network.
Table 3. Preparation formulations of highly transparent \( \text{SiO}_2 \) aerogels.

| Samples | TMOS (wt.%) | TMOS (g) | Methanol (g) | \( \text{NH}_4 \text{OH} \) (g) | Deionized Water (g) |
|---------|-------------|----------|--------------|-----------------|---------------------|
| 1       | 15          | 2.47     | 10.4         | 0.019           | 3.6                 |
| 2       | 15          | 2.47     | 10.4         | 0.038           |                     |
| 3       | 25          | 4.67     | 10.4         | 0.077           | 3.6                 |
| 4       | 25          | 4.67     | 10.4         | 0.058           |                     |
| 5       | 35          | 7.53     | 10.4         | 0.097           | 3.6                 |
| 6       | 35          | 7.53     | 10.4         | 0.038           |                     |
| 7       | 45          | 11.45    | 10.4         | 0.097           |                     |
| 8       | 45          | 11.45    | 10.4         | 0.058           |                     |
| 9       | 45          | 11.45    | 10.4         | 0.038           |                     |

Figure 11. Pictures of \( \text{SiO}_2 \) aerogels with different transparency: (a) sample 12 and (b) sample 20.

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