

## Research Article

# A Novel Environmental Route to Ambient Pressure Dried Thermal Insulating Silica Aerogel via Recycled Coal Gangue

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Coal gangue, one of the main hazardous emissions of purifying coal from coalmine industry, is rich in silica and alumina. However, the recycling of the waste is normally restricted by less efficient techniques and low attractive output; the utilization of such waste is still staying lower than 15%. In this work, the silica aerogel materials were synthesized by using a precursor extracted from recycled silicon-rich coal gangue, followed by a single-step surface silylation and ambient pressure drying. A low density ( $\sim 0.19 \text{ g/cm}^3$ ) nanostructured aerogel with a 3D open porous microstructure and high surface area ( $\sim 690 \text{ m}^2/\text{g}$ ) was synthesized, which presents a superior thermal insulation performance ( $\sim 26.5 \text{ mW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$  of a plane packed of 4-5 mm granules which was confirmed by transient hot-wire method). This study offers a new facile route to the synthesis of insulating aerogel material by recycling solid waste coal gangue and presents a potential cost reduction of industrial production of silica aerogels.

## 1. Introduction

Silica aerogel is one of the hot topics nowadays, because of its exceptional physical properties, for example, high porosity and specific surface area, low density, and thermal conductivity [1–6]. Amongst all attractive characteristics of the material, the most well-known feature is its superinsulating ( $\lambda \leq 13 \text{ mW}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ ) at the Standard Temperature and Pressure (STP) condition and low flammability [3]. One of the state-of-the-art methods to prepare silica aerogel is sol-gel combined with ambient pressure drying. So far, organic silicon monomers, such as tetramethylorthosilicate (TMOS), tetraethyl orthosilicate (TEOS), and polyethoxydisiloxane (PEDS), are generally selected as precursors [4, 7, 8] and the trimethylchlorosilane (TMCS) [9, 10], hexamethyldisiloxane (HMDSO) [11], and hexamethyldisilazane (HMDS) [7, 9, 12] were used for silylating silica gels, and aerogels were dried under ambient pressure conditions.

Coal gangue, a hazardous solid waste of purifying coal from coalmine industry, is rich in silica and alumina. But the recycling of such waste is normally restricted by less efficient techniques and low attractive outputs [13]. Nowadays, with

an ever growing of coalmine industry, 3.8 billion tons' coal gangue was reported in 2014 in China with an increasing rate of 0.2 billion tons per year; it has been recognized as one of the main hazardous industrial emissions [14]. Many efforts have been made to develop facile and effective routes to extend the coal gangue applications; however, the utilization of such waste is still staying lower than 15% [15], and the main applications are limited in typical building materials, for example, concrete filling materials and composite cements [16]; therefore, the overall development of the coal gangue is narrow, and the efficient approach and potential application field studies are of great importance for the resource recovery of waste coal gangue.

Since it is rich in silicon, coal gangue could be used as a cheap silicon raw material. Few literatures have been reported on the relatively pure activated silica and its applications in aerogel materials. Dong et al. [17] extracted the high purity silica aerogel from coal gangue by acid leaching with a yield of 68.04%. Hou et al. [18] have investigated the optimal temperature ( $700^\circ\text{C}$ ) and time (2 hours) for extracting potential available silicon (around 17.65 wt.%). Zhu et al. [19] reported the synthesis of silica aerogels from coal

TABLE 1: Elemental analysis of the raw coal gangue (wt.%).

SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	SO <sub>3</sub>	K <sub>2</sub> O	MgO	P <sub>2</sub> O <sub>5</sub>	MnO	V <sub>2</sub> O <sub>5</sub>	σ <sup>a</sup>	Total
43.59	12.05	10.95	9.01	0.50	19.29	2.31	0.61	0.84	0.05	0.10	±0.03	100

<sup>a</sup>Standard deviation.

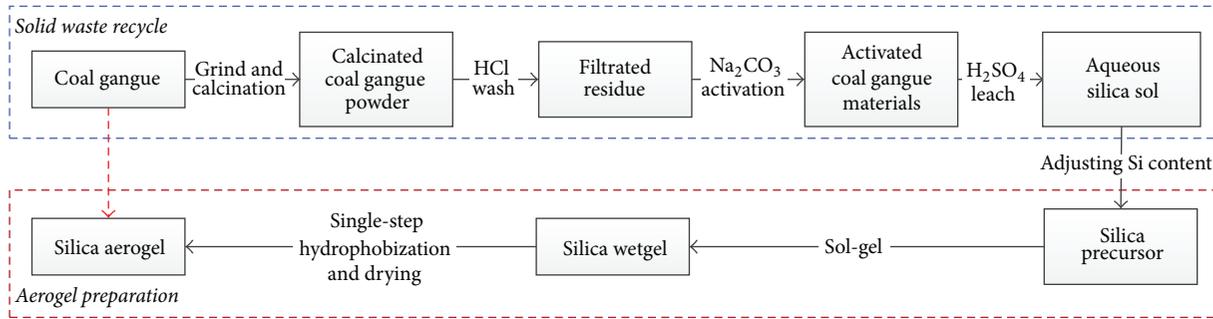


FIGURE 1: Schematic presentation of the main steps of silica aerogel preparation from coal gangue.

gangue by ambient drying of trimethylchlorosilane modified gels; the 88.97% porosity and 0.256 g/cm<sup>3</sup> density were achieved, but there is no report on the extraction yield and thermal performance of the xerogels. Overall, there are few reports on silica xerogel preparation from coal gangue, the reported utilization of the silicon resource was lower than 70%, and no systematical characterizations of such materials, but quite a lot of publications, were reported for precipitated silica fabrications [20–22]; however, the main challenge of aerogel preparation is controlling high SiO<sub>2</sub>/Na<sub>2</sub>O molar ratio in the extracted silicon sol (over 1:1, which needs to restrict Na<sup>+</sup> loading or increase silicon concentration) [23] and high extraction efficiency of silica (which requires high Na<sup>+</sup> concentration and large amount of solvent) and low concentration of impurities, such as Al, Fe, and Mg, in the final gels (which affects the transparency and thermal performance of aerogels). In this case, if we can efficiently wash the impurities out, extract silica, and adjust the SiO<sub>2</sub>/Na<sub>2</sub>O ratio, this will concomitantly ameliorate both the silicon extraction efficiency and final aerogel performances. It has been motivated to develop a feasible route for such high performance materials preparation, that is, silica aerogels and its corresponding composite materials with a great potential of recycle use of low cost and environmental correlated large stocked solid waste.

In this work, waste coal gangue was used as the rich silicon raw material for the preparation of silica aerogel by using a “3-step extraction” route to extract and purify SiO<sub>2</sub> from coal gangue, adjusting SiO<sub>2</sub> concentration, promoting sol-gel transition, and following single-step solvent exchange/hydrophobization/ambient drying; over 96% silicon was extracted as the silica precursor; lightweight (~0.19 g/cm<sup>3</sup>) and nanostructured granular aerogel materials were synthesized by this route (Figure 1), which shows a decent thermal conductance ( $\lambda$ ) of  $26.5 \times 10^{-3} \text{ W}\cdot\text{m}^{-1} \text{ K}^{-1}$  of 4-5 mm packed granules. The study offers a feasible route for synthesizing silica aerogel from recycled waste coal gangue

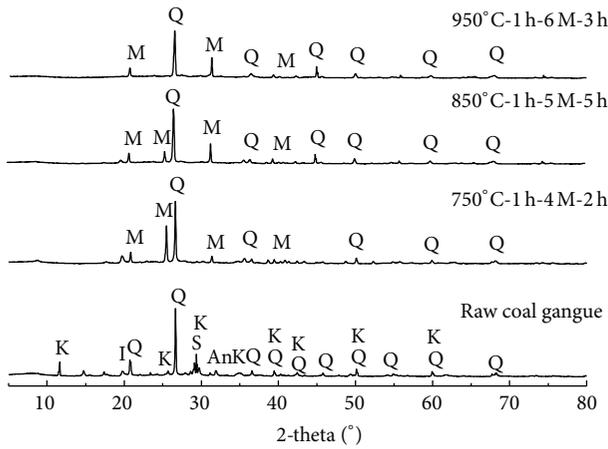
and presents potential niche markets established from such novel route synthesized aerogels.

## 2. Experimental

**2.1. Chemicals.** Hydrochloric acid (HCl, 37%), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>, 98%), and sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>, >99.8%) were purchased from Cancheng Chemical Co., Ltd., China; ammonium hydroxide solution (NH<sub>3</sub>·H<sub>2</sub>O, 28–30%) was obtained from Shanghai Lingche Chemical Co., Ltd., China. Chlorotrimethylsilane (TMCS, 99%, Sigma-Aldrich Co.) was used for the hydrophobization treatment of the aerogels. Ethanol (ACS > 99.8%, Fluka) and heptane (98%, Fluka) were used as the organic solvents. Deionized water was homemade and used in all experiments. All chemicals were used as received without any further purification.

### 2.2. Silicon Precursor Preparation

**2.2.1. Sources of Raw Coal Gangue.** Raw coal gangue was provided by Jintan Coal Mining Co., Ltd., Changzhou, China. The raw materials were crushed by a hammer crusher (PZC-180X150, Aolian Technology Co., Ltd., Changzhou, China) and passed through a 0.3 mm sieve. As-received coal gangue contains less than 15 wt.% C; the elemental and mineral analysis were measured by X-ray fluorescence (XRF) spectrometer and Powder X-ray diffraction (XRD), as shown in Table 1 and Figure 2, respectively. The main crystalline components of the coal gangue could be identified in Figure 2 as illite (X<sub>2</sub>Y<sub>4-6</sub>Z<sub>8</sub>O<sub>20</sub>(OH,F)<sub>4</sub>: X is K and Ca, Y is Al, Mg, and Fe, and Z is Si or Al), kaolinite (Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>), siderite (FeCO<sub>3</sub>), and quartz (SiO<sub>2</sub>) and so forth, which are consistent with the literature [24], but the specific peaks of siderite and illite are relatively weak and mostly overlapped by the peaks of kaolinite and quartz, which are confirmed as the main components of this coal gangue.



Q: quartz  
K: kaolinite  
I: illite  
An: ankerite  
M: mullite  
Ch: chlorite  
S: siderite

FIGURE 2: X-ray diffraction of the raw coal gangue and coal gangue after acid leach and typical chemical formula of the mineral components: quartz ( $\text{SiO}_2$ ), illite ( $\text{KAl}_2[(\text{Al},\text{Si})\text{Si}_3\text{O}_{10}]$ ), kaolinite ( $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ), ankerite ( $\text{CaMg}(\text{CO}_3)_2$ ), mullite ( $\text{Al}_6\text{Si}_2\text{O}_{13}$ ), and siderite ( $\text{FeCO}_3$ ).

**2.2.2. Extraction of  $\text{SiO}_2$  from Coal Gangue.** Around 44 wt.%  $\text{SiO}_2$  in the raw coal gangue is shown in Table 1, which is the target material to be extracted out for using as precursor of silica aerogel preparation.

A 3-step protocol was designed by considering an efficient separation of silicon out of raw coal gangue material, as follows:

- (i) leaching out impurities, that is,  $\text{Fe}^{2+}$ ,  $\text{Al}^{3+}$  (partially),  $\text{K}^+$ ,  $\text{SO}_3$ , and so forth, by treating the coal gangue in a muffle furnace (SX2-2.5-12, Shanghai Hede Instrument Co., Ltd., China) and fast cooling it down in a cool water to activate the raw materials by introducing crystalline defects and keeping reactive amorphous structures: the impurity ions, for example,  $\text{Al}^{3+}$  and  $\text{Fe}^{3+}$ , could be extracted for industrial use as well [17, 25], which is not the main topic and will not be discussed in this paper. The optimized calcination process is  $850^\circ\text{C}$  which was determined by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) [24]. After calcination, the coal gangue was leached by 5 mol/L HCl solution for 5 hours [24],
- (ii) transferring stable silicon-rich components, such as kaolinite and quartz, to easily soluble compounds, for example, albite and nephelite: in order to improve the  $\text{SiO}_2$  extraction efficiency [24], as-treated coal gangue materials were mixed with  $\text{Na}_2\text{CO}_3$  in the respect mass ratio of 0.8, 0.9, 1.0, 1.2, 1.3, 1.4, 1.5, and 1.6 and calcinated at  $750^\circ\text{C}$ ,  $800^\circ\text{C}$ ,  $850^\circ\text{C}$ , and  $900^\circ\text{C}$  for 1, 2, 3, 4, and 5 hours,
- (iii) extracting silicon by using relative high concentration  $\text{H}_2\text{SO}_4$  solution: the activated coal gangue was

dispersed into 4 mol/L  $\text{H}_2\text{SO}_4$  solutions at room temperature and the solid-liquid weight ratio was kept as 1:10.

### 2.3. $\text{SiO}_2$ Aerogel Synthesis

**2.3.1.  $\text{SiO}_2$  Wetgel Preparation.** As-prepared silica sols with certain silicon concentration were used directly as aerogel precursor. The resulting aqueous silica sol has a pH of 0.5, the sol is getting viscous after the addition of sulfuric acid, the gelation time depends on the concentration of  $\text{H}_2\text{SO}_4$ , and the gelation occurred about 10 minutes when 4 M  $\text{H}_2\text{SO}_4$  was added.

**2.3.2. Surface Modification and Ambient Drying.** After gelation, the gel was washed 3 times in the diluted 1M  $\text{H}_2\text{SO}_4$  solution at  $55^\circ\text{C}$  for 24 hrs. After aging, hydrophobization of the gel surfaces (chemical modification with trimethylsilyl (TMS) groups) was carried out by soaking the gels in a heptane/ethanol/TMCS solution with a molar ratio of 15:1:1 at  $65^\circ\text{C}$  for 24 hrs. The gels were then dried in a convective oven at  $120^\circ\text{C}$  for 2 hours.

### 2.4. Characterization

**Mineral and Elemental Analysis.** X-ray diffraction (XRD) analysis was recorded on APEX II DUO X-ray system (Rigaku Corporation, Japan) equipped with a  $\text{Cu K}\alpha$  source at 40 mA and 40 kV. The data were collected from a 2-theta degree in the range of  $10^\circ$  to  $80^\circ$  at a step of  $6^\circ/\text{min}$ . DTA and DSC were performed on a thermogravimetric analyzer (TGA92, SETARAM, France) in the air atmosphere. 10 mg sample was performed with the heating rate of  $10^\circ\text{C}/\text{min}$ . X-ray fluorescence (XRF) spectrometer was analyzed on EA2400II system (PerkinElmer, USA) at 50 kV and 40 mA.

**Scanning Electron Microscope (SEM) Characterization.** Silica aerogel materials were analyzed by coating them with a 10 nm thick platinum layer. SEM analysis of all materials was performed on FEI Nova NanoSEM 230 instrument (FEI, Hillsboro, Oregon, USA) at an accelerating voltage of 10 kV and a working distance of 5 mm.

**Transmittance Electron Microscope (TEM) Characterization.** The microstructure of the aerogel was recorded on a transmission electron microscopy (Philips-EM 420) operated at an accelerating voltage of 200 kV.

**Thermal Conductivity.** The thermal conductivity was measured on an in-house built transient hot-wire device at  $25^\circ\text{C}$  and 50% relative humidity (RH) [24]. The corresponding results were analyzed by the method published by Karaipeki et al. [26]. The Cu/Ni alloy wire was chosen as 73 mm in length and 0.127 mm in diameter to obtain the optimum ratio of 575 [27]. The transient lambda was calculated by

$$\lambda = \frac{VI/4\pi L}{dT/d(\ln t)}, \quad (1)$$

TABLE 2: Acid leach experiment of the coal gangue.

Sample	Calcination temperature (°C)	Calcination time (h)	HCl concentration (mol/L)	Reaction period (h)	Silicon concentration in the residue (%)
CSi-1	750	1	4	2	72.84 ± 1.50
CSi-2	750	2	5	3	79.06 ± 1.13
CSi-3	750	4	6	5	79.10 ± 2.55
CSi-4	850	1	5	5	79.03 ± 0.81
CSi-5	850	2	6	2	78.37 ± 3.10
CSi-6	850	4	4	3	75.45 ± 1.72
CSi-7	950	1	6	3	68.88 ± 2.31
CSi-8	950	2	4	5	74.43 ± 1.55
CSi-9	950	4	5	2	68.69 ± 2.10

where  $V$  and  $I$  are the voltage and current, whose fixed values are 1 V and 0.23 A in this experiment,  $L$  is the length of CuNi alloy wire (73 mm, 0.15  $\Omega$  mm<sup>2</sup>/m), and  $dT/d(\ln t)$  is the average fitting slope of the measurements.

*Evaluation of Density and Porosity.* The bulk density was determined from the envelope volume, which is recorded on GeoPyc 1360, Micromeritics, US, and the skeletal density is measured on AccuPyc 1340, Micromeritics, US. The porosity of the aerogels ( $\epsilon\%$ ) was calculated using the following:

$$\epsilon\% = \left(1 - \frac{\rho_{\text{bulk}}}{\rho_{\text{skeleton}}}\right) \times 100, \quad (2)$$

where  $\rho_{\text{bulk}}$  and  $\rho_{\text{skeleton}}$  are the volumetric mass densities of aerogel and skeletal density of silica, respectively.

*Evaluation of the Brunauer-Emmett-Teller (BET) Specific Surface Area.* Nitrogen adsorption and desorption isotherms at  $-196^\circ\text{C}$  were obtained on Micromeritics ASAP 2020a instrument. The specific surface area of the samples was determined by the Brunauer-Emmett-Teller (BET) method. The pore size distribution was obtained from the desorption branch of the isotherm using the Barrett-Joyner-Halender (BJH) model. The pore volume ( $V_{\text{pore}}$ ) and average pore diameter ( $D_{\text{pore}}$ ) were calculated from the density of the aerogel and its specific surface area using the following:

$$V_{\text{pore}} = \frac{1}{\rho} - \frac{1}{\rho_{\text{skeleton}}}, \quad (3)$$

$$D_{\text{pore}} = \frac{4V_{\text{pore}}}{S_{\text{BET}}}, \quad (4)$$

where  $\rho$  is the density of the aerogel,  $\rho_{\text{skeleton}}$  is the density of aerogel skeleton, and  $S_{\text{BET}}$  is the specific surface area as measured using BET.

### 3. Results and Discussion

*3.1. Removal of the Soluble  $\text{Fe}^{3+}$  and  $\text{Al}^{3+}$  Impurities.* In order to extract relative pure silica out of coal gangue for fabricating aerogel materials, a calcination and HCl leaching

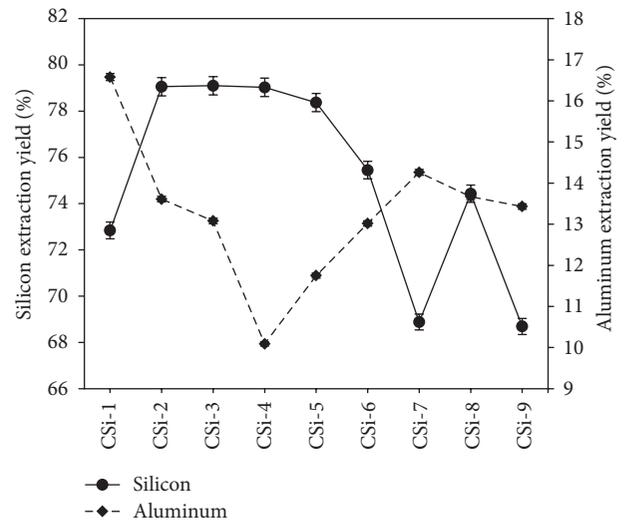


FIGURE 3: The concentration of silicon and aluminum after HCl leach.

process was designed to remove most of soluble impurities (namely, some  $\text{Fe}^{3+}$  and  $\text{Al}^{3+}$  compounds and  $\text{K}^+$  and  $\text{P}^{5+}$  oxides), listed in Table 2. After heat treating and HCl leaching, most of the quartz phase remained, the peaks assigned to kaolinite were no longer distinguished anymore, as shown in Figure 2, which correlates with a degradation of Al/Si compounds, the peaks belonging to iron mineral of siderite were getting weaker, especially at  $750^\circ\text{C}$  and  $850^\circ\text{C}$  treatments, which indicates an effective removal of Fe and Al compounds from the coal gangue, and most of the silicon phases were still retained. After acid leaching, most samples show relatively high silicon contents over 74 wt.%; according to Figure 3, the most attractive protocol is that the coal gangue was treated at  $850^\circ\text{C}$  for 2 hours and then leached by 5 mol/L HCl solution for 5 hours; the sample CSi-4 keeps relatively high concentration of silicon and removes most of aluminum.

*3.2. Chemical Activation of Coal Gangue with an Addition of  $\text{Na}_2\text{CO}_3$ .* The most promising coal gangue intermediate CSi-4 was further activated by the addition of  $\text{Na}_2\text{CO}_3$ ;



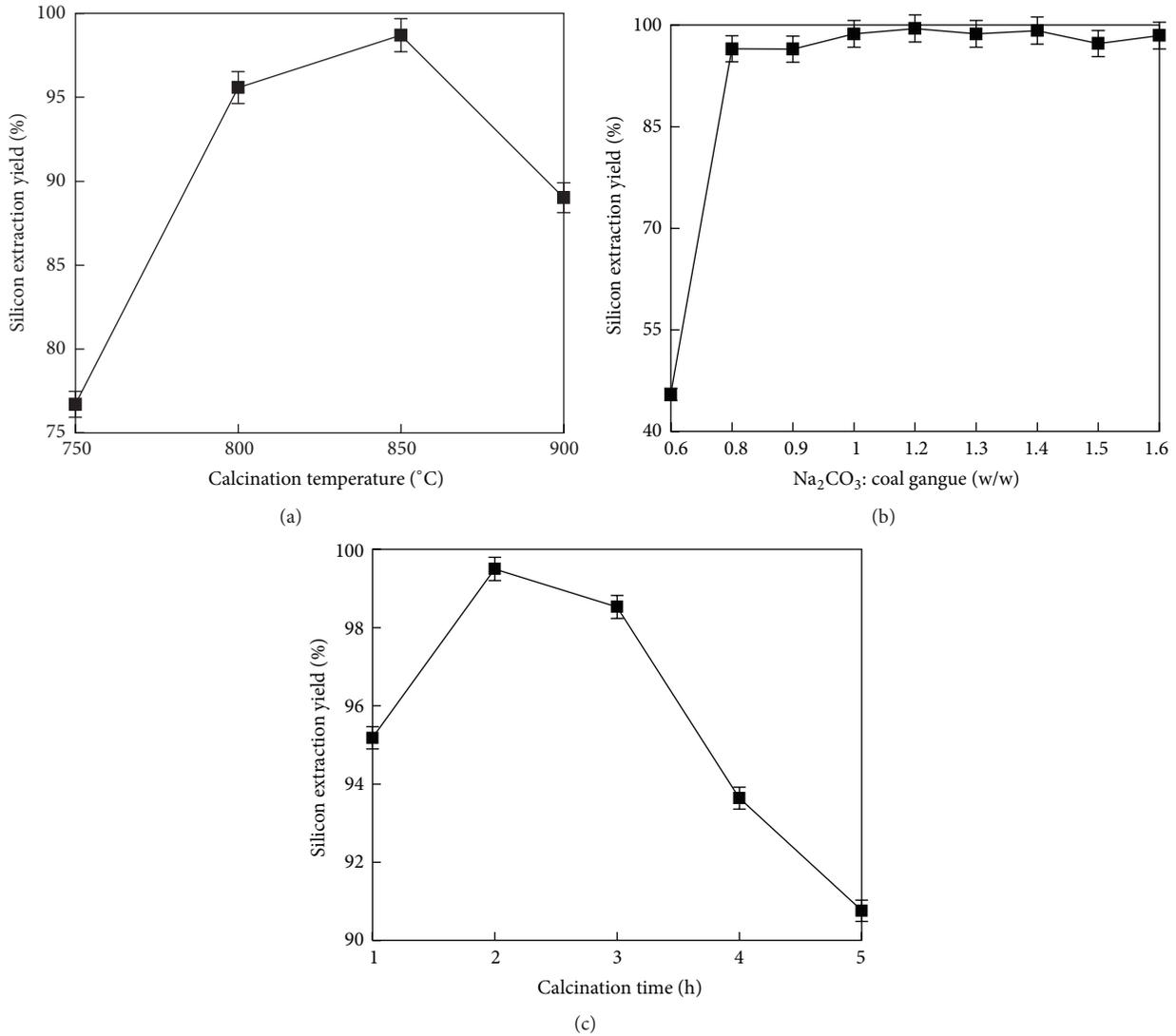


FIGURE 5: The silicon extraction yields adjusted by (a) the calcination temperature, (b) the Na<sub>2</sub>CO<sub>3</sub> to coal gangue ratio, and (c) calcination time.

SEM (Figure 6). Homogenously distributed silica building blocks (around 20–50 nm) were assembled to form a three-dimensional network with the pores ranging from several nm to over 100 nm.

A detailed examination of the aerogel materials under TEM (Figure 7) verifies a hierarchical porous structure with a typical pearl necklace structure with most of the primary colloidal particles in a range of 5–8 nm and 1–2 nm interparticle pores, which is in agreement with the reported typical waterglass-based silica aerogel structures [28]. The hierarchical structure of the aerogel materials observed from the microscopic analysis is mainly attributed to the different size of “building blocks” of the gel particles; the TEM shows around 5 nm primary SiO<sub>2</sub> particles; the primary particles were agglomerated to build the secondary clusters and shown as the blackberry structure in the SEM images. The varied sizes of the particles and pores lead to a superporous light

TABLE 4: Physical properties of the silica aerogel.

$\rho_{\text{bulk}}$ (g cm <sup>-3</sup> )	0.19 ± 0.05
$\rho_{\text{skeleton}}$ (g cm <sup>-3</sup> )	2.21 ± 0.02
$\epsilon\%$	91.4%
$S_{\text{BET}}$ (m <sup>2</sup> g <sup>-1</sup> )	690
$V_{\text{pore}}$ (cm <sup>3</sup> g <sup>-1</sup> )	4.81
$D_{\text{pore}}$ (nm)	27.5

gel structure (Table 4) and show a successful preparation of aerogel materials from the coal gangue raw material.

The N<sub>2</sub> adsorption-desorption isotherms and relevant pore size distribution of the aerogel prepared from CSi-4-1 are shown in Figure 8. The BET measurement could be influenced equilibrium intervals of the gas adsorption [28].

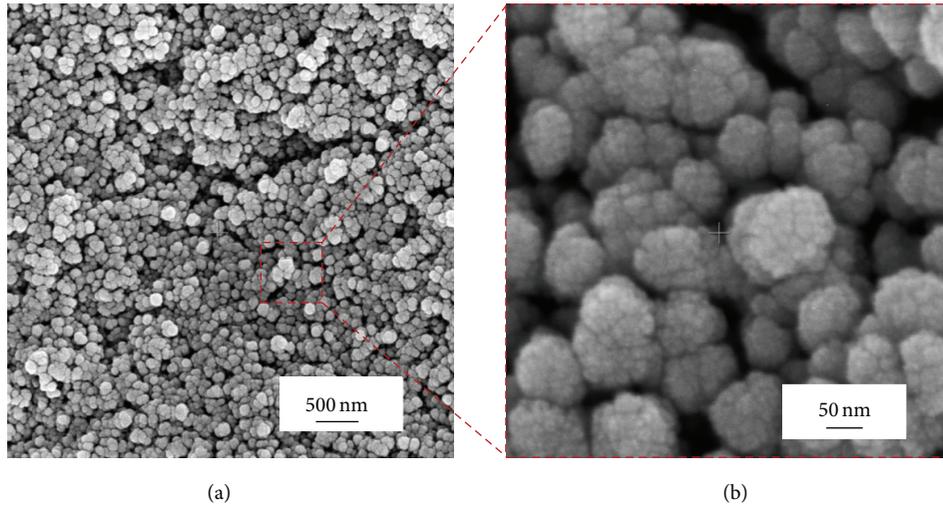


FIGURE 6: SEM images of aerogel materials derived from CSi-4-1 at (a) 50,000x and (b) 400,000x magnifications.

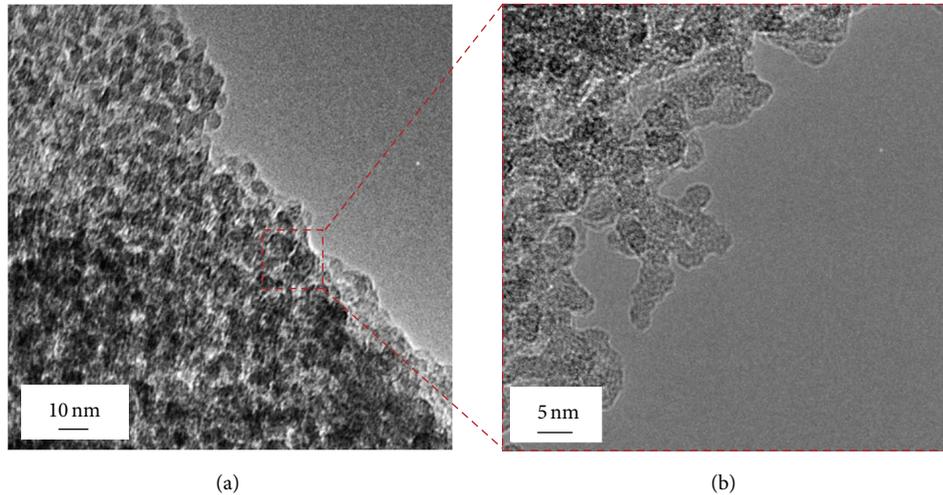


FIGURE 7: TEM images of aerogel materials.

Therefore, in this paper, we tried to repeat the measurements twice with different equilibration times, and the deviations of the main parameters (e.g., surface area, pore size, and pore volume) were around 5%. The isotherm of such aerogel material is close to type 4. The majority of the uptake takes place between relative pressures of 0.1 and 0.95, and the pore volume  $V_{\text{pore}} = 4.81 \text{ cm}^3 \text{ g}^{-1}$  and pore size  $D_{\text{pore}} = 27.5 \text{ nm}$  were calculated from density and surface area, as shown in Table 4, in agreement with the microscopic analysis of Figures 6 and 7, in which the majority of species of mesopores (2–50 nm) were observed.

The Barret-Joyner-Halenda (BJH) equilibrium model was applied for pore size analysis. The pore size distribution (PSD) analysis of silica aerogel shows a minority of micropores ( $\sim 15\text{--}25 \text{ nm}$ ) and a small amount of macropores (from 50 nm all the way up to 55 nm) were identified. The BJH pore size analysis is mostly valid for the pore size in the range of  $\sim 1$  to 150 nm (mesopores and part of macroporous regime);

therefore, some of the big pores built by the secondary clusters (shown in SEM, Figure 6) are not detectable. But the pore size and surface area of the coal gangue derived aerogel are comparable to the typical aerogels prepared from TEOS and waterglass [28]; in this case, it could be assumed that the large macropores are the minority species and indicate that the aerogel materials prepared from coal gangue present similar structures as typical silica aerogel [29] and should display identical performance, that is, thermal insulation, to such material.

**3.4. Thermal Conductivity of Coal Gangue Derived Silica Aerogels.** Thermal insulation is one of the main applications of aerogel-like materials. Ambient thermal conductivity of the granular coal gangue derived aerogel was measured on a packed  $1 \times 1 \times 10 \text{ cm}^3$  granule filled frame by transient hot-wire method [24]; a thermal conductivity of  $26.5 \times$

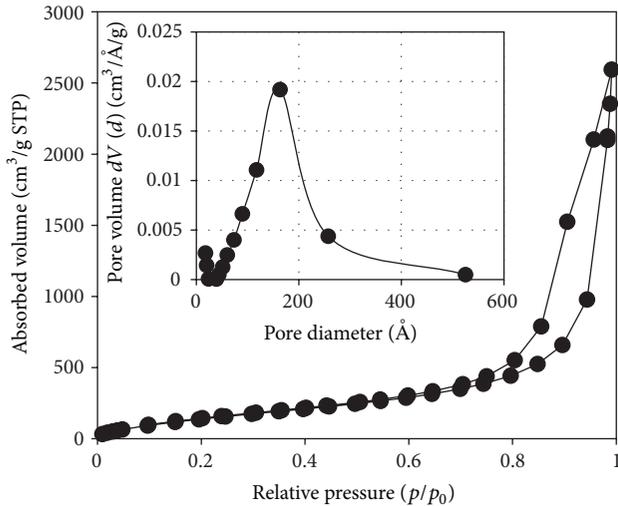


FIGURE 8:  $N_2$  adsorption-desorption (BET isotherm) and BJH pore size distribution (inset, derived from absorption branch) of the aerogel materials.

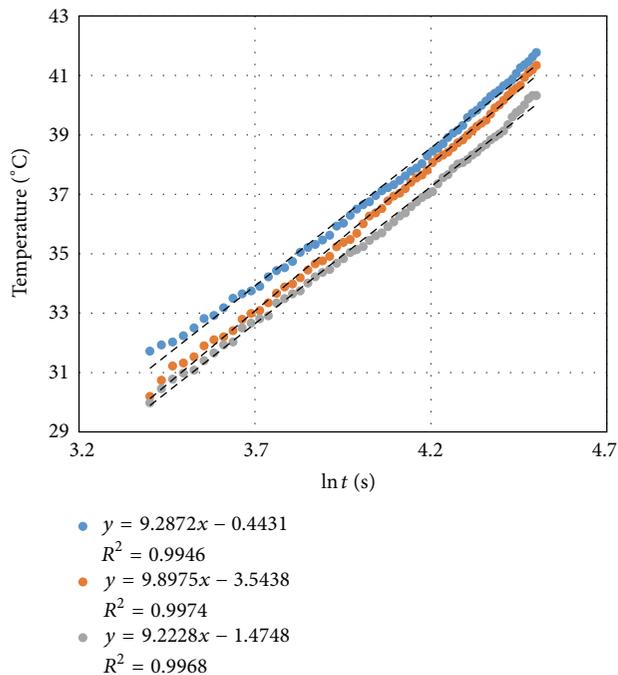


FIGURE 9: Evaluation of thermal conductivities of aerogel material by transient hot-wire method.

$10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$  was calculated from (1) by using the slopes of Figure 9. The in-house measurement device was calibrated by the standard material (Expanded Polystyrene Board) with known thermal conductivity and measurements were repeated 3 times with the deviation of 3.9%. A slight higher value (typical silica aerogel thermal conductance of  $12\text{--}18 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$ ) [28] is probably due to a high bulk density of the gel and loosely packed aerogel granule during measurement [30].

As compared to the density dependent thermal conductivity of typical silica aerogels prepared from TEOS,  $18 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$  at  $0.2 \text{ g cm}^{-3}$  [7], coal gangue derived silica aerogel displays higher thermal conductance, which is close to the reported waterglass-based aerogels [31].

#### 4. Conclusion

We have developed a 3-step (acid purification,  $\text{Na}_2\text{CO}_3$  activation, and acid leaching) route for effective extraction of silicon from a recycled coal gangue; an extraction yield of 99.5% w/w can be reached. After sol-gel transition, the wetgel was directly modified by TMCS/EtOH/n-heptane in single-step solvent exchange/silylation for fabricating mesoporous silica aerogel material. Typical hierarchical mesoporous structure was confirmed by SEM, TEM, and BET analysis, the pore size ranges from several nm to around 100 nm, and most of the pores are in the mesoporous domain (20–50 nm). The aerogel density is  $0.19 \text{ g cm}^{-3}$  with high specific surface area ( $690 \text{ m}^2/\text{g}$ ) and high porosity (>90%). The final aerogel material presents a good thermal insulation property of  $26.5 \times 10^{-3} \text{ W m}^{-1} \text{ K}^{-1}$ , which was limited by in-house designed hot-wire transient method and may be improved in the future by applying an optimized granule packaging method.

This work demonstrates a feasible and economic ambient route to thermal insulating silica aerogels from the recycled solid waste, which offers insights into a cost reduction for industrial production of silica aerogels.

#### Competing Interests

The authors declare that they have no competing interests.

#### Authors' Contributions

Pinghua Zhu and Meng Zheng contributed equally to this work.

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