



A novel low-cost method of silica aerogel fabrication using fly ash and trona ore with ambient pressure drying technique



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ABSTRACT

Highly porous and hydrophobic silica aerogel is fabricated using fly ash and trona ore as the starting materials and the cost-effective ambient pressure drying technique. The optimal calcination parameters are determined as temperature of 850 °C, holding time of 2 h, and the trona ore/fly ash mass ratio of 1.4. The CO₂ release mechanism during calcination is proposed and has been verified by the Thermogravimetric and Differential Scanning Calorimetry analysis (TG/DSC) and X-ray fluorescence (XRF) technique. The hydrogel is derived from reacting the calcination mixture with sulphuric acid solution, followed by filtration. The impurities can be effectively removed through water washing and solvent exchange processes. No ion exchange resin is used in this preparation method, and thus it is a safe, inexpensive and much more straightforward process. In order to minimize drying shrinkage, the hydrogel is first transformed into an alcogel by soaking in ethanol, after which the alcogel is surface modified with the hexane/ethanol/trimethylchlorosilane (hexane/ethanol/TMCS) mixture solution. It has been observed that the specific surface area, BJH desorption pore volume, and BJH desorption average pore diameter all first increase and then decrease so that reach to the maximum values once the heat treatment temperature approaches 500 °C due to the oxidation of —CH₃ groups on the silica skeleton surface. The final product presents special characteristics including: (1) high thermal stability for maintaining hydrophobicity up to 476 °C, (2) contact angle of the as-dried silica aerogel as high as 151°, (3) silica particle size of ca. 3–6 nm, agreeing well with the model as reported, (4) and the 500 °C heat treated sample possessing a large specific surface area of 856.2 m²/g, a large pore volume of 2.92 cm³/g, and a BJH desorption average pore diameter of 17.1 nm. The proposed inexpensive approach produces silica aerogel with superior properties and is a scalable-manufacturing method for large-scale industrial production.

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1. Introduction

Aerogels are light-weight materials that are dried in such a way to avoid the skeleton collapse and leave the air inside the intact nanostructures. Aerogels generally have low thermal conductivity, low bulk density, high specific surface area, low refractive index, low sound velocity and high optical transmission [1–3]. Owing to these unique properties, aerogels find applications in a number of fields such as thermal insulations, acoustics, chemical sensors, catalyst barriers, particle detectors, adsorbents and electronics. Among all kinds of aerogels, silica aerogel has attracted great attention and has been studied for >80 years. However, silica aerogel faces a problem of hydrophilicity under humid environment due to —OH on the surface that is caused by partially hydrolyzed alkoxy residues. Silica aerogels easily deteriorate with

time and thus their use is limited due to their sensitivity to atmospheric moisture and water [4]. Therefore, increasing the hydrophobicity of silica aerogel via introducing non-polar groups (Si-R, where R represents Vinyl, Alkyl or Aryl groups) has been investigated over the past years [5–7]. It is found that this process often associates with surface modification on the wet gel skeleton, which is also a part of the cost-effective ambient pressure drying (APD) technique [2,6–7]. Silica aerogels are conventionally fabricated by the supercritical drying method (SCD). The products via this method have high purity, good stability and high porosity. However, SCD is always involved with high temperature and pressure, which causes operation risk and triggers relatively high cost because of the use of expensive organic silicon alkoxides (TMOS, TEOS). In addition, SCD method always costs much due to the adoption and maintenance of the SCD apparatus. Therefore, APD method has superior advantages over SCD method for silica aerogel fabrication in considering the fabrication cost and safety issues.

Except for these two issues that SCD technique brings, the high cost of starting materials has also restricted silica aerogel's large-scale

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fabrication [8]. Therefore, researchers have attempted to use no or low-cost solid wastes and abundant natural resources for aerogel fabrication in recent years. This route of high-value aerogel production using a large amount of tailings and wastes, including, fly ash, coal gangue, kaolin, oil shale ash, rice husk ash, etc. instead of using critical natural resources is of great significance in considering both environment protection and energy saving.

In previous research, the method to fabricating aerogels from wastes is called the solid-liquid reaction route [9–10]. During this route, the wastes are generally pretreated with high temperature to get rid of the impurities, followed by mixing with the alkaline solution, forming water glass, another conventional precursor for fabricating silica aerogels [11]. The ion exchange resin is then used to remove the Na^+ inside, and then ammonia or NaOH solution is used for gelation, thus obtaining the silica wet gel. Aging, solvent exchange, surface modification and drying are then carried out to form the resulting silica aerogels.

Fly ash is a kind of solid waste that is released by factories and thermal power plants, which contains mainly of silica-alumina mixture with little amount of iron, sodium, calcium and potassium [12]. As much as about 800 million tons fly ash has been generated in the world every year [13]. The disposal of fly ash has caused significant economic and environmental problems over the past years [14]. However, current use of fly ash mainly focuses on concrete, bricks, adsorbents, insulation panels and agricultural fertilizer. There lacks of high-value applications of fly ash. Some literatures have been published for aerogel fabrication using fly ash as the starting material via the solid-liquid reaction route [3,15]. In addition, it has been also proved that fly ash can be used for alumina extraction via calcination with solid alkaline at elevated temperatures [16]. Hu [17] presented that kaolin can be activated by adding analytical Na_2CO_3 at elevated temperatures. Trona ore is a kind of abundant and inexpensive natural resource with the main composition of sodium carbonate. Especially for Sweetwater County, Wyoming, which is a major contributor of trona to the world and supplies about 90% of the nation's soda ash [18]. Therefore, it is a feasible way to develop aerogel using fly ash and trona ore as the starting material.

Herein, we present a novel solid-solid route combining with the cost-effective APD for synthesis of hydrophobic silica aerogel using industrial waste-fly ash and abundant natural resource trona ore as the starting materials. The newly proposed method uses large amount of abundant waste and natural resource instead of the conventional and expensive silica resource (TEOS and TMOS), and it avoids the usage of ion exchange resin which will decrease the complexity and total cost of fabrication. The resulting silica aerogel has large specific surface area and high thermal stability of hydrophobicity. The calcination temperatures on the physicochemical properties of resulting silica aerogels are also investigated in this paper.

2. Experimental section

2.1. Materials and synthesis methods

The fly ash was from Wyoming, USA, which was firstly dried at 50 °C, and grounded to pass the 150 μm sieve, with the oxide composition listed in Table 1. The fly ash contains 52.4% silica, 26.5% alumina and small amount of other oxides, such as Fe_2O_3 , CaO, TiO_2 , etc. Trona ore was from Sweetwater county of Wyoming, USA, which was mainly composed of Na_2CO_3 with the purity of 85% (Table 2). Sulphuric acid (H_2SO_4), trimethyl chlorosilane (TMCS), n-hexane, ethanol (EtOH), and deionized water (H_2O) were purchased from Sigma-Aldrich

corporation, USA. All of them were of analytical reagent grade and no further purification was carried out.

The specific preparation route of silica aerogel was shown in Fig. 1. Fly ash and trona ore were mixed after screening the grounded materials to a mesh of 100 using agate mortar. Then, the mixture was put into an muffle, and after that different calcination temperatures (750 °C–950 °C) with varying holding time lengths (1 h–5 h) were carried out on different samples. The calcining mixture was then immersed in 1 M H_2SO_4 solution with a certain ratio of 8 mL/g. The silica sol was then obtained after vacuum filtration, with the residues for further characterization. The silica sol with a certain volume was then put into the oven, and gelation occurred after ca. 12 h under 60 °C. The hydrogel was aged for 1 d, after which 1 M H_2SO_4 solution wash was performed to remove the impurities inside the skeleton at room temperature. The hydrogel was then immersed into EtOH for solvent exchange at room temperature, after which hexane was used to substitute the ethanol inside the pores. Surface modification was then carried out by soaking the wet gel in a mixture with a volume ratio of hexane:TMCS:EtOH = 8:2:1 for 24 h at 40 °C. After a complete surface modification, the silica wet gel was suspended in a liquor and bailed out. The modified silica wet gel was then dried under room temperature for 3 h, and then it was heated at 100 °C and 200 °C for 2 h, respectively, to obtain the super hydrophobic silica aerogel.

2.2. Characterizations

The X-ray diffraction (XRD) patterns were recorded using a Rigaku Smartlab X-ray diffraction analysis with $\text{CuK}\alpha 1$ radiation ($\lambda = 0.15406$ nm). The X-ray tube was operated at 40 kV and 40 mA. Thermal gravimetric analysis (TGA) and differential scanning calorimetry analysis (DSC) were performed by SDT Q600 Thermogravimetric analyzer under flowing air with a constant flowrate (100 mL/min) at a heating rate of 10 °C/min to 1000 °C and 800 °C, respectively. Scanning electron microscopy (SEM) equipped with an energy dispersive spectrum (EDS) was conducted using a FEI Quanta FEG 450 Scanning Electron Microscope. Transmission electron microscope (TEM) and selected area electron diffraction (SAED) images of the aerogels were taken with the FEI Technai G2 F20 S-Twin. The specific surface area, pore volume and pore distribution were measured using a QuadraSorb SI surface area and pore distribution analyzer. The FTIR spectra were taken with a Nicolet iS50 FT-IR with a Smart Orbit Diamond ATR attachment using a resolution of 4 cm^{-1} and 32 scans. The X-ray photoelectric spectroscopy (XPS) measurement was carried out using the ThermoScientific ESCALAB 250 instrument. The contact angle was tested using a contact angle meter (Rame-hart instrument, USA).

3. Results and discussion

Fig. 2 shows the photographs of fly ash, trona ore, calcining mixture and the resulting silica aerogel. The fly ash and trona ore exhibit grey color and light-yellow color, respectively. The mixture derived from calcining fly ash and trona ore at elevated temperature shows a light-green color, which indicates that some impurities, such as iron ion contained in the mixture. The solution also presents light-green color after immersing the mixture into H_2SO_4 solution, however, these impurities can be removed by water wash and solvent exchange process afterwards. Therefore, the resulting silica aerogel shows a semitransparent white appearance after the APD process.

Table 1

Chemical composition of fly ash used for preparing silica aerogel.

Elements	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	SO_3	TiO_2	MnO	P_2O_5	SrO	BaO	LOI
(wt%)	52.4	26.5	5.58	7.95	3.00	0.15	0.73	1.04	0.68	0.10	0.49	0.09	0.21	1.02

Note: LOI refers to the ignition loss of fly ash.

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