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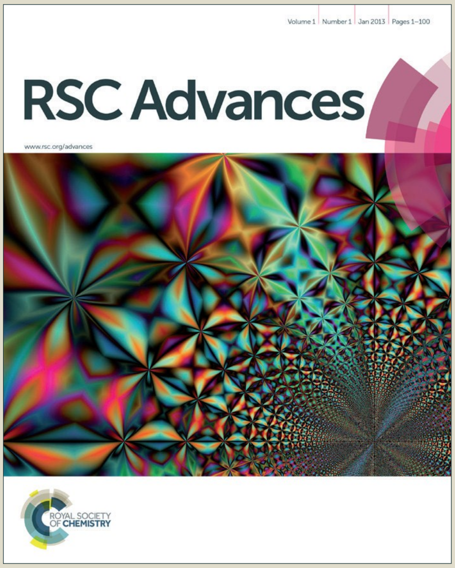

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Facile preparation of SiO_2-Al_2O_3 aerogel using coal gangue as a raw material via ambient pressure drying method and its application in organic solvent adsorption

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Abstract:

Because of the outstanding properties such as large specific area and Brønsted acidity, SiO_2-Al_2O_3 aerogel has been applied in plenty of fields such as adsorption, catalysis, synthetic chemistry. Regarding the preparation of SiO_2-Al_2O_3 aerogel, however, expensive raw materials are always used. In addition, the traditional preparation method is supercritical drying, which has complexity and safety problem. Here, we successfully prepare SiO_2-Al_2O_3 aerogel utilizing coal gangue as a raw material, which is a zero-cost mining waste. More important, ambient pressure drying used in this work overcomes the drawbacks of supercritical drying. Characterizations show that specific surface area of the SiO_2-Al_2O_3 aerogel prepared from coal gangue can be comparable to that of those prepared from relative costly raw materials such as $Al(NO_3)_3{\cdot}9H_2O$ or Al isopropoxide, and tetraethoxysilane with supercritical drying condition. In addition, adsorption tests show that the SiO_2-Al_2O_3 aerogel have good adsorbability of organic solvent. Our work provides an effective route to massively synthesize SiO_2-Al_2O_3 aerogel and opens a new direction of comprehensive utilization

of coal gangue.

Key words: SiO_2-Al_2O_3 aerogel, ambient pressure drying, organic solvent absorption, coal gangue

1. Introduction

After the discovery of the M41S family, mesoporous materials have attracted a large number of investigations because of their outstanding properties.[1-5] Aerogel, a typical mesoporous materials, is a kind of nanoporous material with continuous random network structure.[6] Because of the excellent properties such as the high porosity, high specific surface area and low density, aerogel has attracted a growing number of studies and has received a wide range of applications such as adsorption, catalysis, environmental purification and biochemistry.[7-12] As an unique kind of aerogel, SiO_2-Al_2O_3 aerogel shows strong Brønsted acidity and is a promising absorption and catalysis materials, which is different from pure aerogel such as silica aerogel and alumina aerogel.[13, 14] However, the production of SiO_2-Al_2O_3 aerogel has been restricted because of the expensive precursors, such as $Al(NO_3)_3 \cdot 9H_2O$ and tetraethoxysilane.[15, 16] Therefore, it is highly desirable for finding a kind of cheap material to synthesize SiO_2-Al_2O_3 aerogel.

It is well known that coal gangue is one of the major solid wastes generated in the process of coal mining and washing.[17, 18] Coal gangue accounts for 10%-20% of the original coal production and the production is also increasing every year with a high speed in China.[19, 20] Lager accumulative stockpile of coal gangue not only occupies farmland but also contaminates groundwater, soil, and atmosphere if it is not treated timely and properly.[21, 22] By far, the comprehensive utilization of coal gangue is mainly focused on fundamental application, such as power generation,[23, 24] building material,[25] pavement,[26, 27] and so on. Here, the coal gangue is adopted as starting materials for synthesizing SiO_2-Al_2O_3 aerogel, which not only realizes the SiO_2-Al_2O_3 aerogel using cheap material, but provides a new direction of comprehensive utilization of coal gangue.

In addition, drying method is very important in the synthesis of aerogel. Usually, three drying methods can be used: supercritical drying, freezing drying and ambient pressure drying.[28, 29] Supercritical drying method can obtain aerogel with uniform particle distribution, high porosity and tiny shrinkage, however, issues including complicated processing, safety problem, high cost and equipment requirement restrict its application.[30] As a facile method, freezing-drying method can obtain monolithic and high-quality aerogel. However, the gel network may eventually be destroyed by the nucleation and growth of solvent crystals, which tend to produce very large pores.[31] Besides, time-consuming process makes its commercial application, unreality. Compared to the mentioned drying methods, ambient pressure drying is a facile, relative fast and very cheap way to realize the drying of aerogel.[32] More important, properties of aerogel after ambient pressure drying can be comparable to those prepared using supercritical drying.[33, 34] Herein, a comparable property of SiO_2-Al_2O_3 aerogel is prepared by taking advantage of ambient pressure drying method.

In the current work, the SiO_2-Al_2O_3 aerogel is successfully synthesized by using coal gangue as starting material via ambient pressure drying. To the best of our knowledge, few studies are reported about the synthesis of SiO_2-Al_2O_3 aerogel using coal gangue via ambient pressure drying. In addition, organic solvent absorption of the SiO_2-Al_2O_3 aerogel prepared from coal gangue have also been studied in detail.

2. Experimental

2.1 Materials

Commercial sodium carbonate (Na_2CO_3), hydrochloric acid (HCl), anhydrous ethanol (EtOH), cyclohexane, n-hexane and trimethylchlorosilane (TMCS) were purchased from Beijing, China. All of them are analytical reagent grade and any further purification is not carried out. Coal gangue sample used in the experiment was obtained from Shenmu, Xi'an, China. The chemical composition of air dried basis (A_{ad}) of coal gangue was examined through chemical method (Table 1).

Table 1. Chemical composition of A_{ad}^{*} of coal gangue[wt.%]

Composition	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	K_2O	Na_2O	SO_3	P_2O_5	P^{**}
Coal gangue	66.31	21.04	4.08	0.65	1.13	1.31	3.38	1.30	0.30	0.06	20.31%

*: Air dried basis of coal gangue,

**: Ignition loss of coal gangue.

2.2 Methods

The specific preparation procedure of SiO_2-Al_2O_3 aerogel is shown in Fig. 1(a). Firstly, gangue samples and Na_2CO_3 were prepared by screening the ground raw materials to a mesh of 100 respectively. Then, coal gangue was mixed with Na_2CO_3 at different weight ratio and calcined in a certain temperature (870~930 °C). Subsequently, the sinter was blended with excess HCl solution (ensure that pH of the solution would be less than 3 for preventing the production of precipitation) and stirred for 5-10 min at 100 °C. After that, the resulted mixture was filtered. The filtrate was precisely silica-alumina sol, which could be demonstrated by Tyndall phenomenon (Fig. 1(b)) and insoluble solid was dried (the weight of the insoluble solid is set as m'). Silica-alumina sol was kept motionless to gelatin at 80 °C. After gelation, the wet gel was aged for 3 h at 50 °C to strengthen the gel network. The aged gel was then washed with distill water until the production color was transformed from yellow to none (Fig. 1(c) and (d)), which indicated that $FeCl_3$ trapped inside the system was eliminated completely. Next, SiO_2-Al_2O_3 gel was immersed into EtOH for solvent exchange at 50 °C for 24h. Surface modification was following carried out by soaking the gel in a mixture of TMCS: hexane with a volume ratio of 1:1 for 24h. After entire surface modification, SiO_2-Al_2O_3 gel was suspended in the liquor and bailed out. The modified SiO_2-Al_2O_3 gel was dried at ambient pressure for 24h and then heated at 100 °C, 200 °C for 1h respectively. The final product SiO_2-Al_2O_3 aerogel can be obtained after cooling the oven to room temperature.

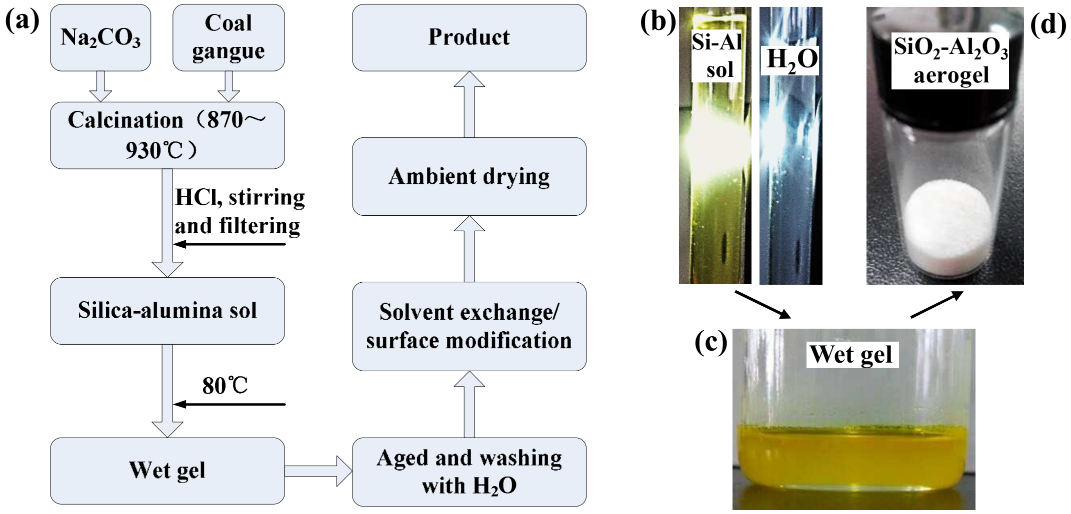

Fig. 1. (a) Schematic diagram of preparation of SiO_2-Al_2O_3 aerogel using coal gangue as the raw materials, (b) pictures of Tyndall phenomenon of silica-alumina (Si-Al) sol by comparing with water, (c) wet gel, and (d) SiO_2-Al_2O_3 aerogel.

For the absorption test of organic solvent, 0.1g SiO_2-Al_2O_3 aerogel was added to the mixture solution of EtOH and cyclohexane (10ml) with the volume ratio of 1:1. 20 minutes later, the resulting aerogel was fetched and weighted.

2.3 Calculation and characterization

The decomposition rate of coal gangue (DG) and organic solvent adsorption rate of the SiO_2-Al_2O_3 aerogel (AR) were calculated by the following equations:

$$\text{DG}=\frac{m'}{m\times(1-p)}\times 100\% \qquad (1)$$

$$\text{AR}=\frac{m_2-m_1}{m_1}\times 100\% \qquad (2)$$

where m is the original weight of coal gangue, m_1 is set as 0.1 g in the current work, m_2 is the weight of the SiO_2-Al_2O_3 aerogel after absorbing the organic solvent, m' is the weight of the insoluble solid from the reaction production of the sinter (coal gangue and Na_2CO_3) and HCl solution (see the Method section in 2.1), and P is the ignition loss of coal gangue.

Density of SiO_2-Al_2O_3 aerogel is determined by Equation (3) as follows:

$$\rho=\frac{M}{V} \qquad (3)$$

where ρ, M, V are the density, weight and volume of SiO_2-Al_2O_3 aerogel, respectively. For the specific procedure, we first screened SiO_2-Al_2O_3 aerogel with a sieve of 200 mesh. Then, the sieved SiO_2-Al_2O_3 aerogel was accurately weighted (M). Next, we loaded the weighted SiO_2-Al_2O_3 aerogel into 5ml measuring cylinder. After shaking the measuring cylinder for 550 times, volume of SiO_2-Al_2O_3 aerogel can be obtained (V). The experiment was done for three times and ρ is the average value from the three experiments.

Various experimental tools was performed to characterize the as-prepared SiO_2-Al_2O_3 aerogel. Transmission electron microscope (TEM) measurement was performed on a Tecnai G2 F20 S-TWIN apparatus. Image of field emission scanning electron microscope (FESEM) was taken on a FESEM (Quanta 600 FEG). X-ray scattering pattern was obtained by using X-ray diffractometer (XRD) with Cu Ka radiation at 0.145nm. Specific surface area data was collected with ASAP 2020 HD88 by using nitrogen adsorption and desorption. The hydrophobicity was measured using hanging drop method on optical contact angle & interface tension meter (SL200KB). The thermal stability was examined by thermogravimetric differential scanning calorimetry thermal analysis (TG-DSC) under air atmosphere with the heating speed of 10 °C /min from room temperature to 900 °C. Spectrum of fourier transform infrared spectroscopy (FTIR) was carried out on on a Bruker Tensor 27 spectrometer with wavenumber range of 4000–400 cm^{-1} at a resolution of 4 cm^{-1} by averaging over 100 scans.

3. Results and discussions

3.1 Effects of different factors on DG

To obtain the high decomposition rate of coal gangue (DG), three experimental parameters are studied, including weight ratio of coal gangue to Na_2CO_3 (m(Coal gangue)/m(Na_2CO_3)), holding time, and calcination temperature. The first investigation factor affecting DG is m(Coal gangue)/m(Na_2CO_3) with keeping holding time 2h and temperature 900 °C (Fig. 2(a)). Reactions happened at high temperature are as follows:

$$3Al_2O_3\cdot 2SiO_2 + 3Na_2CO_3 = 2NaAlSiO_4 + 4NaAlO_2 + 3CO_2 \qquad (4)$$

$$SiO_2(liq) + NaAlO_2 = NaAlSiO_4 \qquad (5)$$

$$Al_2O_3(liq) + Na_2CO_3 = 2NaAlO_2 + CO_2 \qquad (6)$$

$$Al_2O_3(liq) + 2SiO_2(liq) + Na_2CO_3 = 2NaAlSiO_4 + CO_2 \qquad (7)$$

These reactions testify that Na_2CO_3 mainly reacts with Al_2O_3 and SiO_2 inside coal gangue. The main product is $NaAlSiO_4$ that is easily dissolved by HCl solution, which causes the decomposition of coal gangue. Fig. 2(a) demonstrates that DG increases first and then reduces along with the decrement of m(Coal gangue)/m(Na_2CO_3). When m(Coal gangue)/m(Na_2CO_3) is 1:0.6, DG attains the maximum (72.5%). Therefore, from the point of weight ratio of coal gangue to Na_2CO_3, 1:0.6 is optimized to gain high DG.

In addition, the experimental parameter holding time is studied. As can be seen from Fig. 2 (b), DG increases along with the increment of the holding time when the temperature is set 900 ^{o}C and m(Coal gangue)/m(Na_2CO_3) is set 1:0.6. When the holding time is 3h, DG is 85% and only increases by 0.41% at the sacrifice of extending 1h holding time. It can be seen from Table1 that Al_2O_3 plus SiO_2 account for 87.35% of A_{ad} of coal gangue, so DG will not increase significantly even if prolong holding time again. That is to say, almost all SiO_2 and Al_2O_3 have been extracted from coal gangue. Therefore, 3 h is the optimized holding time to obtain high DG.

The third experimental parameter is calcination temperature. With holding time 3h and m(Coal gangue)/m(Na_2CO_3) 1:0.6, effects of calcination temperature on DG are performed. As shown in Fig. 2 (c), the optimized calcination temperature is 900 ^{o}C to obtain higher DG.

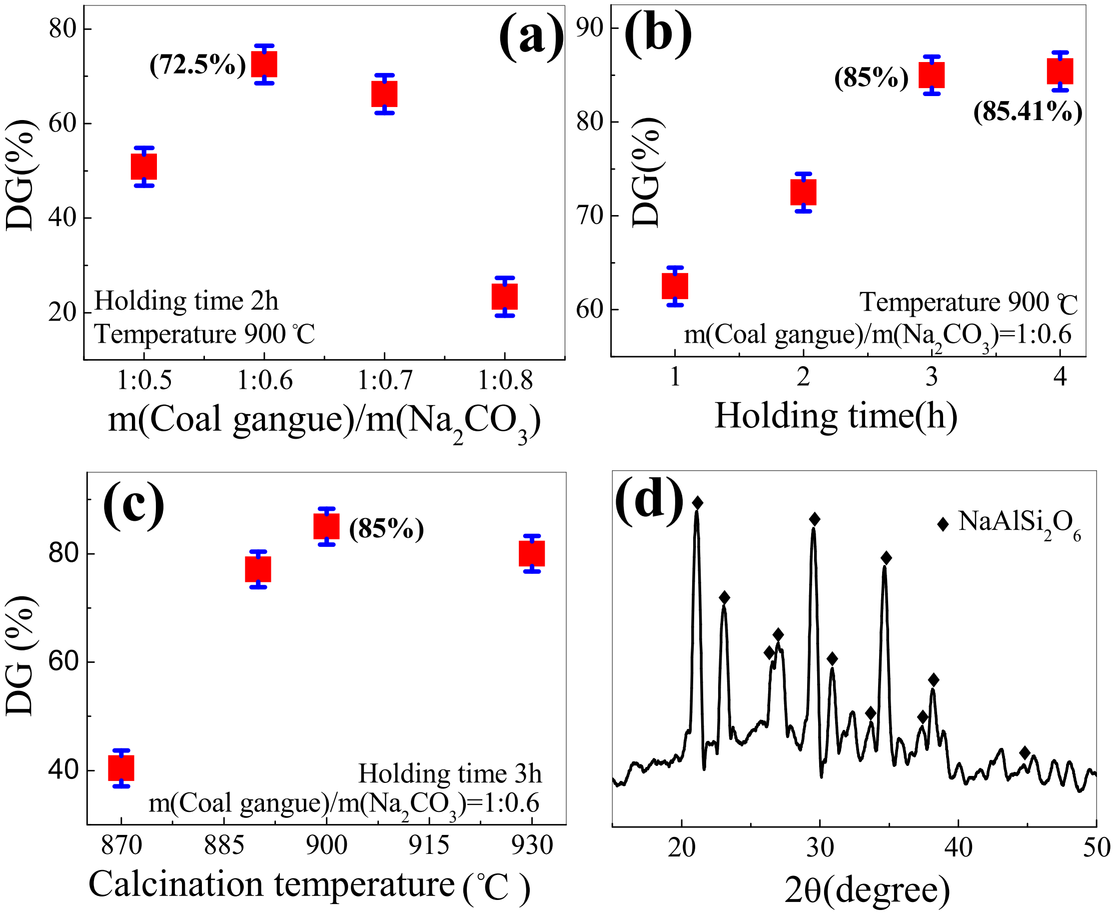

Fig. 2. Effects of different factors on DG: (a) m(Coal gangue)/m(Na_2CO_3), (b) holding time, and (c) calcination temperature. (d) XRD pattern of insoluble substances from reaction of coal gangue and HCl solution.

XRD is carried out to identify the crystal structure of insoluble impurities from the reaction production of calcination mixture and HCl solution. As illustrated in Fig. 2(d), the XRD pattern is precisely match with $NaAlSi_2O_6$ (standard pdf card 46-0012). That is to say, at high temperature, the reaction of coal gangue with Na_2CO_3 produces $NaAlSi_2O_6$ (Equation (8)). Unlike $NaAlSiO_4$ (Equation (4), (5), (7)), $NaAlSi_2O_6$ cannot react with acid. Thus, the decline of DG is caused.

$$Al_2O_3(liq) + 4SiO_2(liq) + Na_2CO_3 = 2NaAlSi_2O_6 + CO_2 \qquad (8)$$

In summary, the optimized parameters to make DG maximize are as follows: m(Coal gangue)/m(Na_2CO_3) 1:0.6, holding time 3h, and calcination temperature 900 ^oC.

3.2 Characteristics of SiO_2-Al_2O_3 aerogel

The final product are synthesized by using ambient pressure drying method under the optimized parameters. Firstly, to study the crystal structure of the final products, the XRD are used. According to the XRD pattern (Fig. 3(a)), strong diffuse peak around 25° of 2θ is due to amorphous SiO_2. As observed between 30° and 80° of 2θ in Fig. 3(a), the XRD pattern is precisely match with Al_2O_3 (standard pdf card 75-0278). Therefore, the XRD result demonstrates that the as-prepared product is SiO_2-Al_2O_3 aerogel. Furthermore, the morphology of the SiO_2-Al_2O_3 aerogel has been studied. Fig. 3(b) is the SEM image of the final product. As can be seen from the image, the sample mainly consists of nanoparticles. Particles of the sample are uniformly distributed and the size is approximately less than 50nm. Importantly, inter-particle connection forms clusters, and then the clusters are linked together with each other. As a result, a large quantity of pores are formed because of the continuous inter-particle connection, as shown in the TEM image in Fig. 3(c). It is noted to say that the ambient pressure drying method are adopted in the preparation of SiO_2-Al_2O_3 aerogel. Ambient pressure drying is a facile, relative fast and very cheap way to realize the drying of aerogel. Based on Fig. 3(b) and (c), the pore size and particle distribution of as-prepared SiO_2-Al_2O_3 aerogel using coal gangue as raw materials via ambient pressure drying is comparable to that of SiO_2-Al_2O_3 aerogel prepared using supercritical drying method (Fig. 3(d)), which is complex and high-cost.[35]

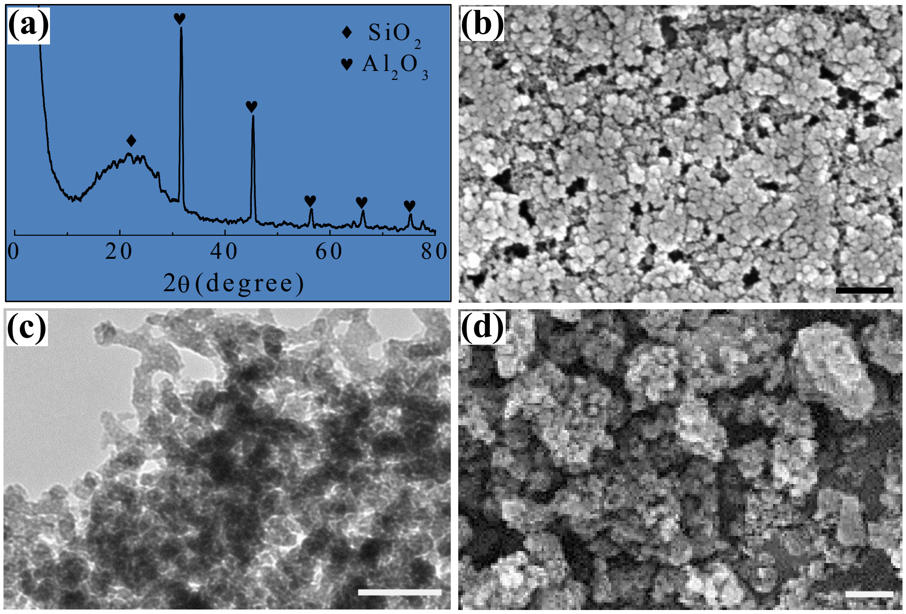

Fig. 3. (a) XRD pattern of the SiO_2-Al_2O_3 aerogel, (b) SEM image of the SiO_2-Al_2O_3 aerogel, and (c) TEM image of the SiO_2-Al_2O_3 aerogel. (d) SEM image SiO_2-Al_2O_3 aerogel prepared using supercritical drying method.[35] All of scale bars are 1μm. Reproduced with permission from ref. 35. Copyright 2005 Elsevier Inc..

3.3 Properties of SiO_2-Al_2O_3 aerogel

Considering the fact that hydrophilic-hydrophobic property plays an important role in organic solvent absorption, the hydrophilic-hydrophobic property is firstly studied. As shown in Fig. 4(a), the contact angle is 134°, which demonstrates that the as-prepared SiO_2-Al_2O_3 aerogel shows strong hydrophobicity. Thus, the SiO_2-Al_2O_3 aerogel has the potential application in organic solvent adsorption. To further clarify the hydrophobicity, the surface functional groups of SiO_2-Al_2O_3 aerogel is studied through FTIR, as shown in Fig. 4(a). Peak around 3480cm^{-1} is attributed to antisymmetry stretching vibration of –OH, which is caused by physical absorption H_2O in air. Peaks at 2960cm^{-1}, 1260cm^{-1} respectively are due to stretching and bending vibration of C-H, which demonstrates that SiO_2-Al_2O_3 aerogel surface is connected with $–CH_3$.[36] The hydrophobicity is caused by $–CH_3$ group connected at SiO_2-Al_2O_3 aerogel. Because of the rotational vibration of H-O-H, absorption peak at 1640cm^{-1} appears.[37] On the other hand, other peaks are also observed. Peaks around 1095cm^{-1} and 460cm^{-1} are ascribed to Si-O-Si bonds. Low intensity of absorption peak of Si-OH near 960cm^{-1} shows that SiO_2-Al_2O_3 aerogel is relatively uniform.[38] More important, peak at 846cm^{-1} is due to the vibration of Al-O bond, which manifests that Si-O-Al bond exists in the sample as well as the successful preparation of SiO_2-Al_2O_3 aerogel using coal gangue.

To study the thermal stability of the SiO_2-Al_2O_3 aerogel, the TG-DSC is performed. Fig. 4(b) is the TG-DSC curve of SiO_2-Al_2O_3 aerogel with the heating rate of 10 ^{o}C/min under atmosphere. From 0 ^{o}C to 431^{o}C, the mass loss is approximate 4%, which is primarily ascribed to the removal of small amount physical absorption water trapped into the sample. Obviously, there is a rapid weight loss (about 4.5%) at 431 ^{o}C, which is attributed to the oxidation of $–CH_3$ connected to the surface of SiO_2-Al_2O_3 aerogel. With the increase of temperature, the weight of sample continues to loss. Those mass loss from 431^{o}C to 900 ^{o}C results from the crystallization change of SiO_2. As a result, SiO_2-Al_2O_3 aerogel prepared from coal gangue can maintain up to 431 ^{o}C in air, which implies that the coal gangue based SiO_2-Al_2O_3 aerogel possesses high thermal stability.

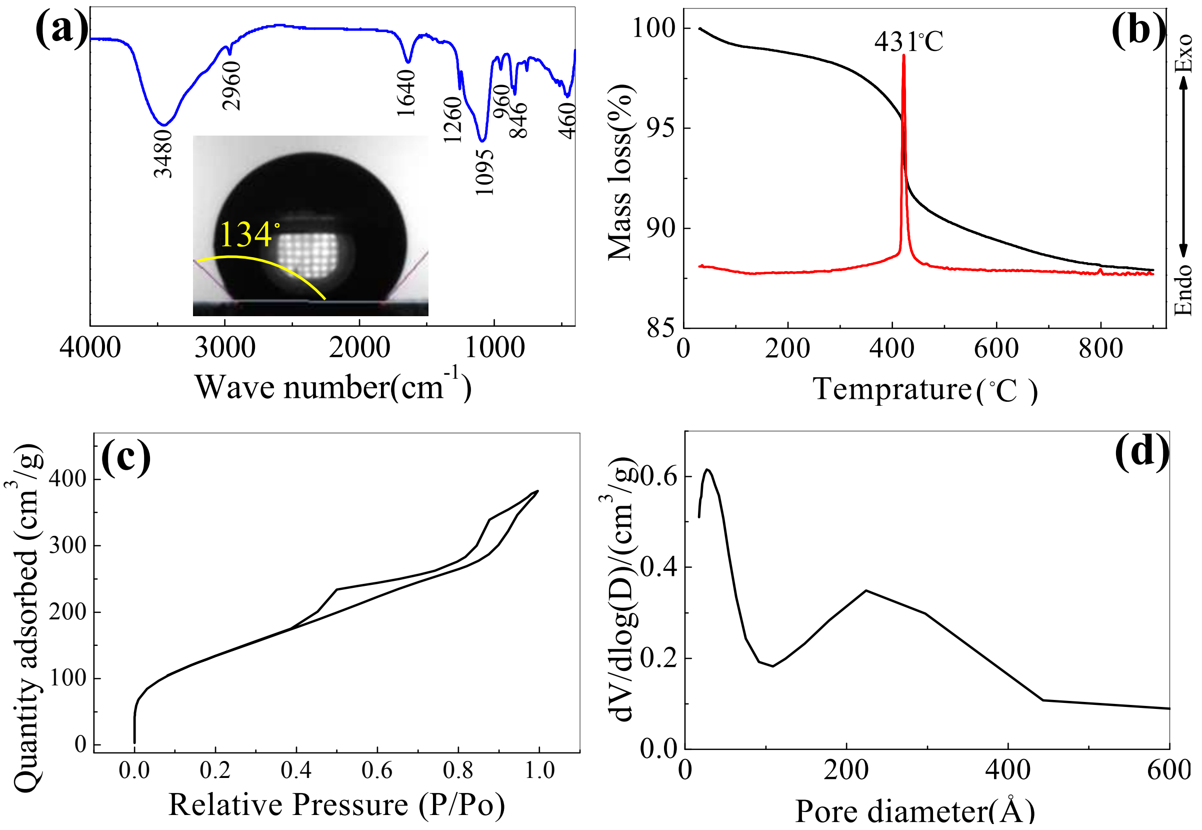

Fig. 4. (a) FTIR spectrum of SiO_2-Al_2O_3 aerogel. Inset of Fig. 4(a) is the contact angle of SiO_2-Al_2O_3 aerogel with water. (b) TG-DSC curve of SiO_2-Al_2O_3 aerogel in air. (c) N_2 adsorption–desorption isotherms of SiO_2-Al_2O_3 aerogel. (d) Pore size distribution of SiO_2-Al_2O_3 aerogel.

Specific surface area also plays a key role in organic solvent absorption. Therefore, the characterization of specific surface area is performed. N_2 adsorption–desorption isotherms and Barrett–Joyner–Halenda (BJH) pore size distribution curve are shown in Fig. 4(c). The isotherm is type IV and the hysteresis loop belongs to H1, which agree well will characteristic of mesoporous materials.[39, 40] The SiO_2-Al_2O_3 aerogel have surface specific area as large as 493.5 m^2/g with the pore volume over 0.59 cm^3/g and the average pore diameter is 4.8 nm. In addition, the calculation result of Equation (3) shows that the density of SiO_2-Al_2O_3 aerogel is 0.3475g/cm^3. This low density of SiO_2-Al_2O_3 aerogel agrees well with large surface specific area. As shown in Fig. 4(d), two pore size distribution ranges are observed: 2-10nm and 10-50nm. The former has overwhelming majority percentage than the latter explaining the small average pore diameter. On one hand, a large number of inter-particle links form small pores that causes the 2-5nm pore size distribution. On the other hand, connections of clusters composed of nanoparticles are inclined to form large pores, which explains the 10-50nm pore size distribution. The above results agree well with the characteristic of TEM image. Therefore, SiO_2-Al_2O_3 aerogel prepared from coal gangue have large specific surface area, small pore size and low density. The specific surface area also can be comparable to SiO_2-Al_2O_3 aerogels prepared under supercritical drying condition. For instance, using supercritical drying condition, C. Hernandez *et al*. obtained SiO_2-Al_2O_3 aerogels with specific surface area range of 253-729 m^2/g.[41]

3.4 Application in absorption of organic solvent

The application of SiO_2-Al_2O_3 aerogel in treatment of organic solvent is one of significant aspects. Its absorption rate of organic solvent (mixture of EtOH and cyclohexane) was examined by being exposed to atmosphere for 0, 10, 20, 30 days. As can be seen from Fig. 5, the fresh produced SiO_2-Al_2O_3 aerogel can absorb organic solvent triple itself in weight, which accords with the BET data. With the extending of time, the adsorption capacity will slightly decline. However, though being exposed in air for 30 days, the absorption rate of SiO_2-Al_2O_3 aerogel of organic solvent only decreases by 40%. Hence, SiO_2-Al_2O_3 aerogel prepared from coal gangue not only exhibit good adsorbability, but can maintain high stability in air, which also agrees with the TG-DSC result.

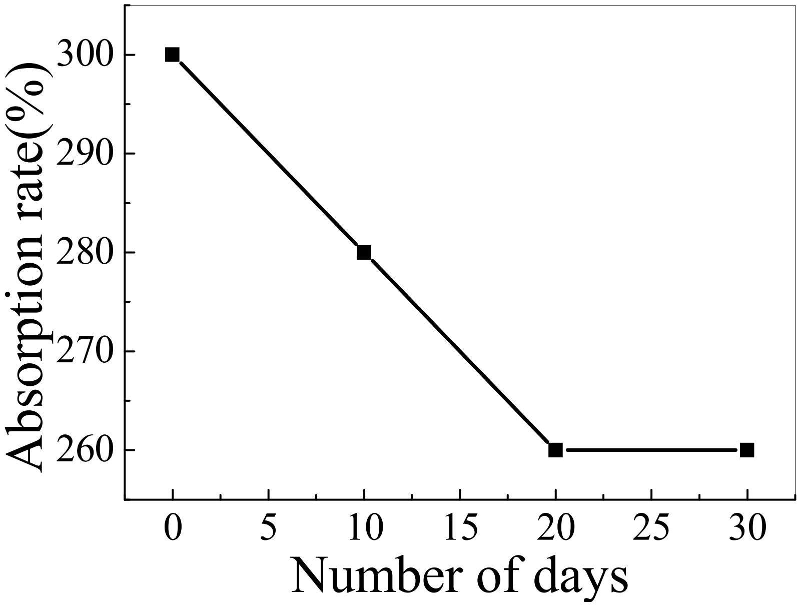

Fig. 5. Absorption rate tendency of SiO_2-Al_2O_3 aerogel to organic solvent with time.

4. Conclusions

In summary, we synthesize SiO_2-Al_2O_3 aerogel using coal gangue as the raw materials via ambient pressure drying. The optimal parameter to obtain the maximum DG is: m(Coal gangue)/m(Na_2CO_3) 1:0.6, holding time 3h, and calcination temperature 900 °C. Under this condition, almost all SiO_2 and Al_2O_3 can be extracted from coal gangue (DG reaches up to 85%). Residue from reaction of HCl solution and calcination mixture are detected by XRD. Result shows that the residue are $NaAlSi_2O_6$, which cannot react with acid and should be responsible for the decrease of DG. More important, the final product have strong hydrophobicity (contact angle with water 134°), high thermal stability that cannot be oxided before 431 °C in air, large specific surface area (493.5 m^2/g), small pore size (average pore diameter 4.8nm) and low density (0.3475g/cm^3). In addition, adsorption tests show that the SiO_2-Al_2O_3 aerogel have good adsorbability of organic solvent. Our work can provide an effective route to massively synthesize SiO_2-Al_2O_3 aerogel and open new direction of coal gangue comprehensive utilization.

Acknowledgements

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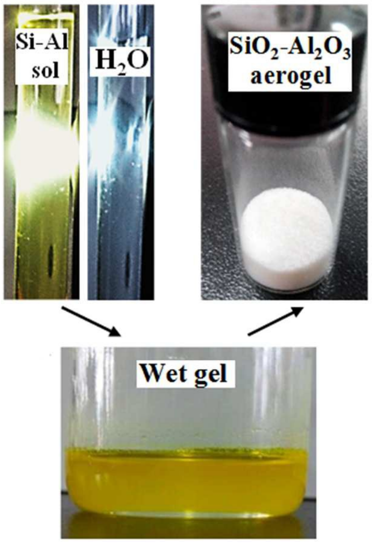
Table of content:



The SiO_2-Al_2O_3 aerogel has been synthesized using coal gangue as a raw material via ambient pressure drying method.