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# FABRICATION AND MICROSTRUCTUAL CHARACTION OF SILICA AEROGEL BY AGING PRESSURIZATION

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## ABSTRACT

$SiO_2$ aerogel with Light-weight and low thermal conductivity is a promising candidate for thermal insulator used for aerospace vehicles. In this paper, we report the preparation and microstructural characterization of $SiO_2$ aerogel by aging pressurization using supercritical drying method. The results showed that the aging pressurization can rapid increase the bulk density from 0.1g/cm$^3$ to 0.45g/cm$^3$ with the pressure changing from 200Pa to 600Pa. When the pressure increases to 800Pa, the density was increased to 0.46g/cm$^3$ slowly. Further polycondensation is driven by the increasing of contact area between skeleton particles when the aging pressure increased. The grid structure became densification and saturation when the aging pressure approached 800Pa. SEM method gives the evidence of increase of aging pressure, which can help to increase the size of secondary grains. Nitrogen sorption-desorption measurements exhibit an unimodal pore distribution and low specific area and porosity with the increase of aging pressure. Real density test showed that the bulk density increased by pressure. Bulk density, gain size and pore structure distribution can be controlled effectively by aging pressurization.

## 1 INTRODUCTION

Silica aerogels have been examined for numerous applications owing to their low density, high porosity, and thermal insulating properties [1]. Silica aerogels were first produced in the 1930s, but saw little development for several decades [2, 3]. Super insulator was put forward strong attract attention at a materials engineering international congress in the 1992, which was defined that the thermal conductivity was lower than that of the air at the same temperature [4]. The porosity is higher than 90% and the very small pores with very high-surface areas (such as from 200 to 1100 m$^2$/g) of the internal structures, which can block the gas thermal conductivity. Air conduction is greatly reduced due to the very fine pore size of the aerogels. At the meantime, these new aerogels are distinguished by their very fine pore sizes (from about 1 to 20 nm), which can produce a low solid- phase thermal conductivity [2]. Then, NASA scientists have found aerogels critical for several space mission, such as Mars Pathfinder mission in 1997, stardust mission in 1999, and so on [5]. Other areas also look favorable: architectural and appliance insulation, shipping containers, refractory insulation, and so on [6-8]. Perhaps the most serious limitation of these materials is their poor mechanical strength [9]. Previous efforts toward reinforcing silica aerogels have been focused on simple gel aging (Ostwald coarsening), the addition of filler materials (typically organic polymers), or surface treatments [10-12]. Aperture distribution directly has an effect on the mechanical and thermal insulation properties. In this paper, we attempt to fabricate the silica aerogel by aging pressurization and to achieve homogeneous aperture.

$SiO_2$ aerogels were prepared by sol-gel method. The sol is prepared by a silica solution (tetraethoxysilane) and by addition of catalyst, gelation is achieved. Then the gel is aged in its mother solution with pressure-plate for 120h, following which the hydrogels were extracted into ethanol. To prevent the collapse of the gel structure, drying is made to take place under supercritical dried conditions using ethanol.

## 2 EXPERIMENTAL

### 2.1 Preparation method

$SiO_2$ aerogels were prepared by sol-gel method. The sol is prepared by a silica solution (tetraethoxysilane) and by addition of catalyst, gelation is achieved. Then the gel is aged in its mother solution with pressure-plate for 120h, following which the hydrogels were extracted into ethanol. To prevent the collapse of the gel structure, drying is made to take place under supercritical dried conditions using ethanol.

### 2.2 Test method

Skeletal density, was determined by helium pycnometry, and nitrogen adsorption/ desorption was used to determine pore size distribution. Scanning Electron Microscopy (SEM) was performed on samples to characterize pore morphology.

## 3 RESULTS

The aging process strengthens the gel, so that shrinkage during the drying step is kept to a minimum.

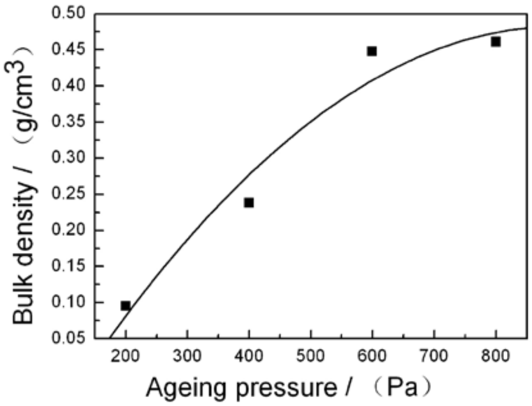


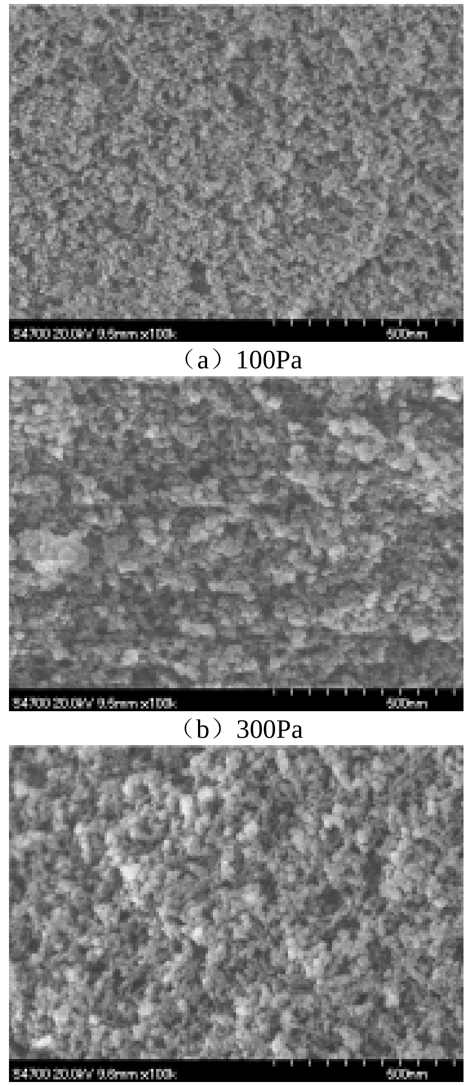

Fig. 1 The relationship of aging pressure and bulk density of silica aerogels

Fig. 1 presents the bulk density increase with the pressure changing from 200Pa to 800 Pa by aging pressurization. When the pressure changed from 200 Pa to 600 Pa, the bulk density increased rapidly. When the pressure continues to 800 Pa, the density increased to $0.46 g/cm^3$ slowly. Aging increases the stiffness and strength of the alcogel by adding newmonomers to the silica network and by improving the degree of siloxane cross linking [2]. The bulk density in line may be estimated as equation (1):

$$y=-0.179+0.00147x-8.125\times10^{-7}x^2 \qquad (1)$$

Where x is the aging pressure, y is the bulk density.

Fig. 2 shows the photographs of the samples prepared by different aging pressure. The grain size is bigger than those samples without pressure. Neck growth from reprecipitation of silica dissolved from particle surface onto necks between particles during the aging process. Dissolution and precipitation of smaller particles change onto larger ones. These two mechanisms will operate simultaneously, but at different rate. When the aging pressure increases from 200Pa to 600Pa, the neck growth from reprecipitation of silica dissolved from particle surface onto necks between particles at a quick rate. At the same time, the aperture distribution becomes homogeneous. When the aging pressure increases to 800Pa, the lager particles were prior precipitated. Gel framework is easy to form strong branch structure crosslinking, which could lead to decrease of specific surface area, as shown in fig. 3. Branch structure further condensation polymerization. On the other hand, distance increment between the grains for the filling the alcohol solvent was solved. Then, grain surface and skeleton linkage area occurred condensation easily, which can raise the strength and hardness of gels.



（a）100Pa

（b）300Pa

（c）700Pa

Fig. 2 SEM photographs of samples prepared by different aging pressurization

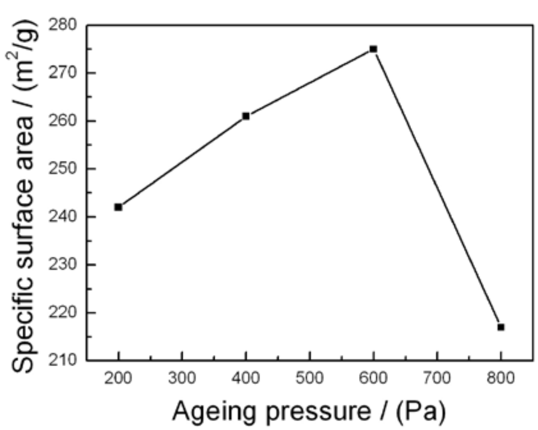



Fig.3 The relationship of aging pressure and specific surface area

Photographs depicting the aperture distribution during aging are provided in fig. 4. Single peak of cumulative pore volume is not obvious without pressure. Though the peak value is higher than those samples with aging pressure, the test result is limit by the nitrogen adsorption/ desorption method (2~50nm). Because smaller aperture is easy to sintering at high temperature could limite their application. Single peak of cumulative pore volume is obvious when the aging pressure added to 400 Pa, which suggests that aperture distribution is more homogeneous. Single peak of cumulative pore volume is about 20 nm and distribution peak is not obvious when the aging pressure is 800 Pa.

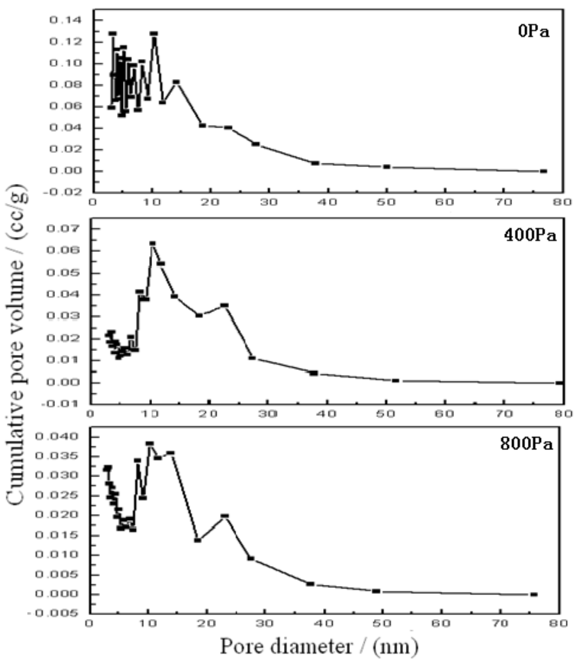



Fig. 4 Aperture distribution at different aging pressure

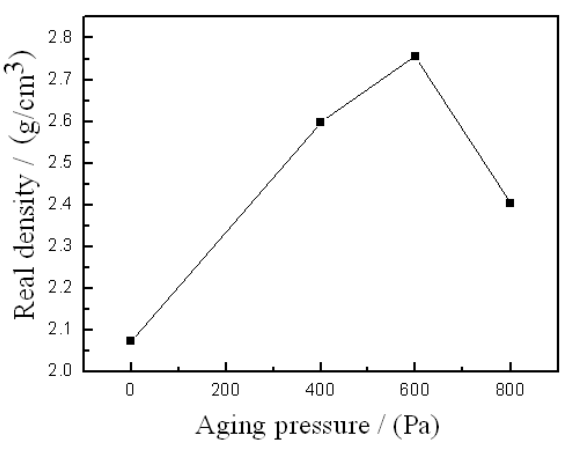



Fig. 5 The relationship of aging pressure and skeleton density of silica aerogels

Fig. 5 shows the relationship of aging pressure and skeleton density. The curve shows an increase with the aging pressure changing from 0 to 600Pa, then decrease with the aging pressure 800Pa. It is different from the law of bulk density. Skeleton density decreases when the aging pressure achieved 800Pa. Skeleton volume should increase, which correspond to the law came from bulk density, SEM photographs and $N_2$ adsoption/desorption test.

## 4 CONCLUSIONS

Silica aerogels were prepared by aging pressurization method. The effection of aging pressure were evaluated by SEM、$N_2$ adsorption-desorption method, skeleton density tests, and so on. Distribution of aperture structure, grain size, skeleton density and bulk density could be adjusted by changing the aging pressure, which can provide data and ideas for structural design. Silicon aerogel is ideal when the pressure is 600Pa.

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