Fabrication and characterization of Aerogel-Polydimethylsiloxane (PDMS) Insulation Film

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Abstract. The building has a large impact on the space heating demand and the indoor environment is affected by climate or daylight. Hence, silica aerogel has generally used as a film to reduce the coefficient of the window in the building. Silica aerogel is a suitable material to apply for insulation material with lower thermal conductivity than that of air to save interior energy. However expensive precursor and drying process were the main issue of the silica aerogel synthesis and practical usage. We attempt to fabricate aerogel insulation film for energy saving through the economic process under ambient pressure. Silica aerogel was synthesized from rice husk ash, which was an agricultural waste to be able to recycle. Taguchi design was used to optimize the parameters (amount of rice husk ash, pH, aging time) controlling the surface area of silica aerogel. The silica aerogel is prepared by sol-gel processing through acidic treatment and aging. The silica aerogel was obtained by modification of silica hydrogel surface and dry at ambient pressure. Finally, aerogel film was respectively fabricated by the different content of aerogel in polydimethylsiloxane (PDMS). Silica aerogel obtained 21 – 24nm average particle size was analyzed by SEM and silica aerogel with high surface area (832.26 m²/g), pore size (3.30nm) was characterized by BET. Then silica Aerogel – PDMS insulation film with thermal conductivity (0.002 W/mK) was analyzed by thermal wave system. The study demonstrates an eco-friendly and low-cost route toward silica – PDMS insulation film with low thermal conductivity (0.002 W/mK).

1. Introduction

The Building window is affected by the demand for space heating energy. The outside temperature is directly transmitted through the window. If this phenomenon persists, it is difficult to maintain the inside temperature. Therefore, energy is continuously consumed to maintain the indoor temperature. Because of this phenomenon, Insulation is needed to reduce the demand for such heating. Typical materials for insulation include glass wool(0.028-0.043W/mK), mineral wool(0.033-0.038W/mK), silica(0.030-0.004W/mK), and perlite(0.030-0.101W/mK). Therefore, silica aerogel is a suitable material to apply for insulation to save interior energy[1]. Silica aerogels are porous materials consisting more than 90% air and 10% solid silica with a cross-linked network.
structure. Silica aerogel have a large surface area, high porosity, low bulk density, low thermal conductivity. However expensive precursor and drying process were the main issue of the silica aerogel synthesis. In this study, we synthesized the silica aerogel from rice husk ash, which was an agricultural waste to be able to recycle using the Taguchi method [2]. Taguchi method is applied in determining to optimize the parameters controlling the surface area of silica aerogel on the fewer experimental results than those required to study the effect of overall input variables [3]. The silica aerogel is prepared by sol-gel processing through acidic treatment and aging. The silica aerogel was obtained by modification of silica hydrogel surface and dry at ambient pressure. To fabricate Aerogel Polydimethylsiloxane (PDMS) Insulation Film, prepared silica aerogel is combined Polydimethylsiloxane (PDMS). Then, we have investigated the specific surface area of silica aerogels through Brunauer-Emmett-Teller and analyzed the thermal conductivity of the film using thermal wave system.

2. Materials and methods
2.1 Materials
The rice husk used by raw material was obtained in Geoje, Korea. Ethyl alcohol 99.9%, Sodium hydroxide beads 97% and n-Hexane 95% were purchased from DAEJUNG. Hydrochloric acid 36% and Sylgard 184A (Polydimethylsiloxane), Sylgard 184B were prepared by SAMCHUN Sewang hitech silicone respectively.

2.2 Preparation of silica aerogel using the rice husk ash
Taguchi design was used to optimize the parameters (amount of rice husk ash, pH, aging time) controlling the surface area of silica aerogel[4]. The rice husk was calcinated at 600 °C for 4 hours to remove the organic matters. Then Calcinated rice husk ash (RHA) and Sodium hydroxide solution was heating 3, 6, 9g of RHA with 90ml of 1M sodium hydroxide solution at 90 °C for 2hour to produce sodium silicate solution, immediately the mixture was filtered to remove the contaminant. For the sol-gel polymerization, hydrochloric acid was added to the filtrated silicate solution until the pH value of the 4, 7, 9. After gelation, The prepared sample was aged for 1, 5, 7days respectively. The aged gel was washed with water for 12 hours at room temperature. The hydrogel was changed into Ethanol to remove the moisture remaining in the hydrogel pore[5]. Then, The silica gel was immersed in n-Hexane and Tetramethylchloricacid (TMCS) mixture (10:1...
Volume) for 24 hours at room temperature [6]. Finally the surface modified silica gel was dried at 60, 80, 120 and 150 °C for 3, 3, 2 and 2 hours as shown in scheme 1.

Table 1 Synthesis condition of silica aerogel using the Taguchi method

| Run no. | RHA (g) | pH | Aging time (days) |
|---------|---------|----|-------------------|
| 1       | 3       | 4  | 1                 |
| 2       | 3       | 7  | 5                 |
| 3       | 3       | 9  | 7                 |
| 4       | 6       | 4  | 5                 |
| 5       | 6       | 7  | 7                 |
| 6       | 6       | 9  | 1                 |
| 7       | 9       | 4  | 7                 |
| 8       | 9       | 7  | 1                 |
| 9       | 9       | 9  | 5                 |

| Code | Factor | Level |
|------|--------|-------|
| A    | RHA (g) | 1 3 9 |
| B    | pH     | 4 7 9 |
| C    | Aging time (day) | 1 5 7 |

2.3 Selection of levels and factors
The rice husk ash is important in synthesis of silica aerogel. The hydrolysis and condensation reactions of the silica aerogel are different depending on the aging time as well as the pH value. The factors are amount of rice husk ash, pH value, and aging time. We chose the each parameters shown in table 1: amount of rice husk 3, 6, 9g; pH value 4, 7, 9; aging time 1, 5, 7 days to confirm the surface area of silica aerogel.

2.4 Fabrication of Aerogel-Polydimethylsiloxane(PDMS) insulation film
The Aerogel-Polydimethylsiloxane(PDMS) insulation film was respectively fabricated by the different content of silica aerogel in polydimethylsiloxane(PDMS). Silica aerogel and polydimethylsiloxane (PDMS) were mixed by Acoustic mixer middle mode for 5 minute. Then,
sonication for 15 minute and spin coating about 2ml aerogel-PDMS solution. Finally, Dry the sample of coated aerogel-PDMS solution at 70°C as shown in scheme2.

3. Results and discussion

Table 2 The surface area and S/N ratio of silica aerogel by each factors

| Run no. | Surface area (m²/g) | Pore Volume (cm³/g) | Pore size (nm) | S/N ratio |
|---------|---------------------|---------------------|----------------|-----------|
| 1       | 803                 | 1.30                | 5.51           | 58.10     |
| 2       | 253                 | 1.71                | 2.30           | 48.05     |
| 3       | 405                 | 1.98                | 16.72          | 52.14     |
| 4       | 649                 | 0.61                | 3.53           | 56.24     |
| 5       | 226                 | 0.91                | 17.82          | 47.08     |
| 6       | 380                 | 1.30                | 11.59          | 51.60     |
| 7       | 657                 | 0.70                | 4.07           | 56.35     |
| 8       | 348                 | 1.94                | 20.79          | 50.85     |
| 9       | 358                 | 1.71                | 18.92          | 51.08     |

3.1 Determination of optimum conditions

We synthesized silica aerogel with a high specific surface area using the Taguchi method. The specific surface area of silica aerogel synthesized under different conditions was measured via Brunauer-Emmett-Teller (Tristar-2030). The table 2 shows the specific surface area, pore volume and pore size of each sample. After that, the value of specific surface area was used to calculate signal-to-noise ratio (S/N). In the Taguchi method, the Signal and noise ratio (S/N) shows the values required for each output characteristic. In this study, The bigger characteristic (specific surface area) is better when the ideal target value is infinity [7]. The signal-to-noise ratio (S/N) is defined as Eq. (1).

\[ S/N = -10 \log \left( \frac{1}{n} \sum_{i=1}^{n} \frac{1}{y_i^2} \right) \] (1)

If the characteristic value is smaller, it is good when ideal target is zero. The Signal and noise ratio (S/N) is calculated by Eq. (2)

\[ S/N = -10 \log \left( \frac{1}{n} \sum_{i=1}^{n} y_i^2 \right) \] (2)

When the bigger characteristic is better, the signal-to-noise ratio (S/N) is calculated by Eq. (3)

\[ S/N = -10 \log(MSD) \] (3)

The MSD is the mean square deviation from the target value of the quality characteristic. Where:

\[ MSD = \left( \frac{1}{y_1^2} + \frac{1}{y_2^2} + \frac{1}{y_3^2} + \cdots \right) / n \] (4)

In MSD, \( y_1 \) and \( y_2 \) are measured results value, the specific surface area corresponds to this experiment.

The signal-to-noise ratio (S/N) values calculated by substituting the data values of the experiment into the equation1 of the bigger characteristics are shown in Table2.

Table 3 Separation of S/N ratio by specific surface area

| Code | Factor               | Average of S/N ratio |
|------|----------------------|----------------------|
| A    | RHA (g)              | 52.76 51.64 52.74    |
| B    | pH                   | 56.90 48.66 51.60    |
| C    | Aging time (day)     | 53.51 51.79 51.95    |

And then, the signal-to-noise ratio (S/N) values obtained in the previous step are separated into the respective levels in order to determine what are the important factors among the variables affecting
the characteristic values in Table 3. For example, signal-to-noise ratio (S/N) values of 1 level on B factor is 58.10, 56.24, 56.35 and the average of signal-to-noise ratio (S/N) is 56.90. After calculating the average value of each signal-to-noise ratio (S/N), its value is indicated in Figure 1 [8]. The B (value of pH) factor with the longest in level 1-3 is the most influential factor in the specific surface area. Therefore, we can confirm the optimal condition of each factor, which is the one with the higher S/N ratio from Figure 1.

![Figure 1](image-url)

**Figure 1** The average of signal-to-noise ratio (S/N) ratio graph for specific surface area

For example, in the case of the B factor, the optimum condition of the B factor is B1 since the signal-to-noise ratio (S/N) ratio of B1 is larger than B2. Consequently, the optimal condition for each factor is A1B1C1. It is necessary to confirm whether the predicted results match the experimental results after the signal-to-noise ratio (S/N) ratio analysis for each factor is completed.

The predicted signal-to-noise ratio (S/N) is defined as Eq. (5)

\[
\text{predicted signal-to-noise ratio (S/N)} = T + (\text{Optimal value of factor } A - \bar{T}) + (\text{Optimal value of factor } B - \bar{T}) + (\text{Optimal value of factor } C - \bar{T}) \tag{5}
\]

Where:

\[
\bar{T} = \frac{\sum_{i=1}^{n} S/N_i}{n} \tag{6}
\]

It was confirmed through Eq. (5) that the predicted value of the signal-to-noise ratio (S/N) ratio was 58.39. The silica aerogel was synthesized again under the optimum conditions. The signal-to-noise (S/N) ratio of synthesized again silica aerogel calculated to be 58.42 as shown in table 4.

| Sample no.          | Optimal condition | Surface area (m²/g) | S/N ratio |
|---------------------|-------------------|---------------------|-----------|
| Predicted value     | A1B1C1            | 803                 | 58.39     |
| Experimental value  | A1B1C1            | 834                 | 58.42     |

As a result, it was confirmed that the predicted value and the result value of the signal-to-noise ratio (S/N) ratio were almost the same value. The surface of silica aerogel powder analyzed by scanning electron microscope (SEM) as shown in figure 2.
3.2 Thermal properties of Aerogel-Polydimethyl siloxane (PDMS) Insulation Film

Table 5: Density, heat capacity, and thermal conductivity of insulation films containing different mass% of silica aerogel powder

|                  | PDMS      | PDMS_0.1wt% silica aerogel | PDMS_0.2wt% silica aerogel | PDMS_0.4wt% silica aerogel |
|------------------|-----------|-----------------------------|-----------------------------|-----------------------------|
| Density (g/cm³)  | 1.095     | 1.064                       | 1.042                       | 1.041                       |
| Heat capacity (J/g°C) | 1.011   | 0.012                       | 0.019                       | 0.020                       |
| Thermal conductivity (W/mK) | 0.150 | 0.065                       | 0.048                       | 0.002                       |

We fabricated an insulation film using high specific surface area silica aerogel prepared by the ta-guchi method. The density and heat capacity of the sample should be analysed to measure the thermal conductivity [9]. Therefore, we measured the density and the heat capacity of the sample containing different amounts of silica [10]. As a result of the measurement, it was confirmed that the density decreases as the silica aerogel content increases in the insulation film. The heat capacity of the insulation film was also confirmed to increase with increasing silica aerogel content. The thermal conductivity of the film without silica aerogel (0 wt%) was measured to be 0.150 W/mK. And thermal conductivity of insulation film containing different silica aerogel contents (0.1, 0.2, 0.4 wt%) was measured by 0.065, 0.048, 0.002 W/mK. As a result of measuring the thermal conductivity, it was confirmed that the thermal conductivity value decreased as the silica aerogel content increased.

4. Conclusions
In this study, we attempt to develop economical and eco-friendly synthesis process of hydrophobic silica aerogel. The silica aerogel synthesized by the rice husk ash which was an agricultural waste to be able to recycle using Taguchi method. The silica aerogel is prepared by sol-gel processing using acidic treatment. It was dried at ambient pressure after modification of silica hydrogel surface. As a result, the silica aerogel having a high specific surface area (834 m²/g) was synthesized under optimized conditions (amount of rice husk ash:3g, value of pH:4, aging time: 1day). The silica aerogel obtained 21 – 24nm average particle size was analyzed by scanning electronic microscope (SEM), silica aerogel with high surface area (832.26 m²/g), pore size (3.30nm) was characterized by Brunauer-Emmett-Teller (BET). Aerogel-Polydimethylsiloxane (PDMS) Insulation Film was respectively fabricated by the different content of optimized condition silica aerogel in polydimethylsiloxane (PDMS) film. The silica Aerogel – PDMS insulation film with low thermal conductivity (0.002 W/mK) was analyzed by thermal wave system. We are fabricating a
heat insulating film with the same effect as a commercially available insulating. Therefore, we are going to carry out experiments to make insulation films to enable mass production in the future.

5. Reference

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