Optimization of the Sound Absorption Coefficient (SAC) from Cellulose–Silica Aerogel Using the Box–Behnken Design

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ABSTRACT: Noise pollution, which has become a major environmental issue in urban areas, can be minimized using acoustic insulation derived from cellulose–silica aerogel. The raw materials required in the process include waste newspaper-based cellulose, geothermal silica, and NaOH/ZnO solution. Therefore, this study investigates the effect of cellulose, silica, and ZnO concentrations on optimizing the sound absorption coefficient (SAC) using the Box–Behnken design (BBD). The results showed that the optimum conditions were obtained at 39.8578 wt % cellulose, 16.5428 wt % silica, and 0.5684 wt % ZnO. The impedance test for the cellulose aerogel and cellulose–silica aerogel showed SAC values of 0.59 and 0.70, respectively, and were characterized by XRD, FTIR, BET–BJH, SEM–EDX, and TG. The results of XRD and FTIR data indicate that the product was cellulose–silica aerogel, and the SEM micrographs showed that silica particles were attached to the fiber surface. Furthermore, type IV isotherms were observed in the cellulose–silica aerogel, typical of mesoporous materials. The presence of silica strengthened the aerogel structure, improved its thermal stability, and increased the surface area but decreased its pore size.

INTRODUCTION

Housing development in urban areas has increased rapidly due to population and infrastructural growth. It generates noise, thereby decreasing the exigencies for citizens’ comfort. Furthermore, noise causes permanent hearing loss, unpleasantness, stress, cardiovascular system disease, diabetes, hypertension, and other health issues in humans. Likewise, noise can be reduced using noise barriers, enclosures, and absorption materials. Acoustic insulation made from petroleum such as fiberglass, polyester, polypropylene, polyurethane foam, and melamine foam is one of the most effective ways to reducing noise. However, these materials are difficult to recycle, and burning them releases carbon dioxide, methane, and nitrous oxide into the atmosphere. Therefore, there is a need for economical and sustainable acoustic insulation.

Biomass such as cellulose matrix is used as an alternative material for acoustic insulation. Meanwhile, cellulose is the most abundant organic polymer on earth, with unique biodegradability, sustainability, eco-compatibility, and recyclability properties. It is derived from waste newspaper, cotton, flax, corn, and rice straw. The waste newspaper contains about 61% cellulose, 16% hemicellulose, and other inorganic fillers. The high cellulose content indicates its potential as an alternative source for cellulose production. However, the organic compound has limited fire thermal resistance. Therefore, a more environmentally friendly cellulose-based material, such as cellulose aerogel, was developed.

Cellulose aerogel is a highly porous material composed of biodegradable fibers with a relatively large surface area of approximately 400 m²/g when supercritically dried and 100–200 m²/g in freeze-drying. However, in a few cases, it has a surface area ranging from 2 to 5 m²/g and a low thermal conductivity between 0.029 and 0.032 W/mK. The mechanical strength of cellulose aerogels manufactured from recycled fibers is lower than that of natural cellulose fibers. Therefore, it is necessary to improve the mechanical strength of recycled cellulose by adding a super insulating material such as silica aerogel.

Silica aerogel is a lightweight, nanostructured, and highly porous material. It has a low thermal conductivity ranging from 0.015 to 0.025 W/mK and ultralow density similar to that of air, which is 0.00129 g/cm³. Therefore, it is used as a catalyst, adsorbent, thermal insulation, and acoustic insulation. Furthermore, silica aerogel consists of more...

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than 90% air and less than 10% solid silica by volume. It is released from the geothermal power plant in Dieng. The geothermal silica is widely used in various applications, such as an adsorbent, chitosan coating, preparation of silicon, synthesis of silica xerogel, synthesis of silica aerogel, synthesis of mesoporous silica, and superhydrophobic coating. Meanwhile, fly ash, bamboo leaves, rice husks, and bagasse were extensively studied for their silica content.

The initial method for preparing cellulose–silica aerogel is dissolution that is performed using cellulose solvents, such as calcium thiocyanate, ammonium thiocyanate, liquid ammonia/ammonium thiocyanate (NH₃/NH₄SCN), and N,N-dimethylacetamide/lithium chloride (DMAc/LiCl). However, these solvents are unaffordable, limited to laboratory scale, and mostly cause serious environmental problems.

Therefore, new eco-friendly cellulose solvents were recently developed, such as the NaOH solution. This solvent system is confined to low-molecular-weight cellulose; hence, some additives such as zinc oxide (ZnO) are added to enhance the solubility of the NaOH solution.

Several studies have been conducted regarding the synthesis of cellulose–silica aerogel using supercritical drying to preserve the porous structure and produce a flexible and translucent cellulose–silica aerogel. However, this process necessitates the use of a high-pressure vessel, which requires expensive equipment. In another study, freeze-drying was employed to synthesize cellulose–silica aerogel. This method can damage the nanostructured gels because it generates crystal growth and development of stress in the pores, possibly leading to fracture of the matrix. Additionally, an extended aging period is necessary to stabilize the gel network, which necessitates using a solvent with a high sublimation pressure and a low expansion coefficient. Therefore, ambient pressure drying is a promising technology used for large-scale industrial applications.

Demilecamps et al. prepared the cellulose–silica composite aerogels by a one-pot synthesis method. The result showed that the gelation time was reduced by adding sodium silicate to the cellulose–NaOH solution even in the existence of ZnO, which is known as an additive that delays cellulose’s gelation. However, the existence of silica decreases the specific surface area. Additionally, He et al. studied the in situ synthesis of cellulose–silica aerogel by one-step impregnation. It was discovered that impregnating the silica particles into the cellulose matrix increases the mechanical properties and specific surface area. Feng et al. analyzed the synthesis of silica–cellulose aerogels from regenerated cellulose fiber and silica precursor methoxytrimethylsilane (MTMS). The results showed good thermal and acoustic insulation properties. The sound absorption coefficients (SACs) of the silica–cellulose aerogels lie between 0.39 and 0.50, which are better than those of cellulose aerogels (0.30–0.40). Furthermore, the synthesis of a silica–cellulose aerogel composite from waste newspaper-based cellulose, silica from geothermal solid waste, and NaOH/PEG solution for acoustic insulation materials has been investigated using a central composite design (CCD) method by Silviana et al. The optimization result showed the optimum point at 1.78 w/v % PEG and 25 wt % cellulose releasing a maximum SAC value of 0.9896.

This study prepared cellulose–silica aerogels using cellulose from waste newspaper, geothermal silica, NaOH/ZnO solution, and ambient pressure drying. The method used to obtain the optimal cellulose, silica, and ZnO concentrations is the Box–Behnken design (BBD), which has fewer experimental trials than other methods. Furthermore, BBD does not combine all factors at the highest or lowest levels at the same time; hence, it is effective in avoiding experiments under extreme conditions, which may lead to undesirable results.

The morphological structure and acoustic insulation properties of the aerogel were further investigated.

## RESULT AND DISCUSSION

### Effect of Cellulose, Silica, and ZnO on the SAC Value

Table 1 shows the design experiment and SAC values.

| run | cellulose | silica | ZnO | SAC  |
|-----|-----------|--------|-----|------|
| 1   | −1        | 0      | −1  | 0.9276 |
| 2   | 0         | 1      | 1   | 0.9589 |
| 3   | 0         | 1      | −1  | 0.9352 |
| 4   | 0         | 0      | 0   | 0.9653 |
| 5   | −1        | 0      | 1   | 0.9666 |
| 6   | 1         | 0      | −1  | 0.9758 |
| 7   | 1         | 0      | 1   | 0.9369 |
| 8   | 1         | 1      | 0   | 0.9596 |
| 9   | 1         | −1     | 0   | 0.9416 |
| 10  | −1        | 1      | 0   | 0.9359 |
| 11  | 0         | −1     | −1  | 0.9374 |
| 12  | 0         | −1     | 1   | 0.9518 |
| 13  | −1        | −1     | 0   | 0.9297 |

### Table 2. ANOVA for the Cellulose–Silica Aerogel

| source | sum of squares | df | mean square | F value | p value |
|--------|----------------|----|-------------|---------|---------|
| model  | 0.0027         | 9  | 0.0003      | 14.39   | 0.0252  |
| A: cellulose | 0.0004 | 1 | 0.0004 | 19.04 | 0.0233 |
| B: silica  | 0.0003 | 1 | 0.0003 | 13.09 | 0.0363 |
| C: ZnO    | 0.0001 | 1 | 0.0001 | 3.09  | 0.1768 |
| AB       | 0.0000 | 1 | 0.0000 | 1.64  | 0.2899 |
| AC       | 0.0014 | 1 | 0.0014 | 66.75 | 0.0038 |
| BC       | 0.0000 | 1 | 0.0000 | 0.8934 | 0.4143 |
| A²        | 0.0003 | 1 | 0.0003 | 12.50 | 0.0385 |
| B²        | 0.0004 | 1 | 0.0004 | 17.78 | 0.0244 |
| C²        | 0.0000 | 1 | 0.0000 | 0.4931 | 0.5331 |
| residual | 0.0001 | 3 | 0.0000 |        |         |
| total cor| 0.0028 | 12 |            |        |         |
| std. dev. | 0.0046 | R² |        | 0.9774 |         |
| mean     | 0.9495 | adjusted R² | 0.9094 |         |         |
| CV %     | 0.4847 | adeq. precision | 12.8331 |         |         |

Specific compositions are listed in Table 6. The optimum result was obtained at a concentration of 35 wt % cellulose, 15 wt % silica, and 1 wt % ZnO with an SAC value of 0.9653. Based on the above, the Design Expert program suggested the quadratic model. Furthermore, the interaction between variables was analyzed using analysis of variance (ANOVA), as shown in Table 2. The parameters F value, p value, R², adjusted R², and adequate precision were used to check the model adequacies. As shown in Table 2, a p value less than 0.05 indicates that the corresponding model terms are significant, including A, B, AC, A², and B². There is only a 2.52% chance of a large F value, and it could occur due to noise in the experiment; hence, the signal-to-noise ratio was measured using adequate precision of...
12.83, which is greater than 4. This shows an adequate signal implying that the model can be used to navigate the design space. Furthermore, the high $R^2$ implies that the model can only explain 97.74% of the total variation data. The adjusted $R^2$ value of 90.94% shows a high significance between the predicted and actual values.

The quadratic model derived from the regression analysis of the experimental data is given below:

$$
SAC = 0.9653 + 0.0071A + 0.0059B + 0.0029C \\
+ 0.0030AB - 0.0188AC - 0.0022BC - 0.0108A^2 \\
- 0.0128B^2 - 0.0021C^2
$$

where $A$, $B$, and $C$ are cellulose, silica, and ZnO (wt %).

Equation 1 shows that the SAC value is directly proportional to the increase in the concentration of cellulose ($A$), silica ($B$), ZnO ($C$), and cellulose–silica interaction ($AB$), corresponding to a positive constant value. Meanwhile, the interaction between cellulose–ZnO ($AC$) and silica–ZnO ($BC$) gave a negative effect on the SAC value.

Figure 1 shows the predicted vs actual values for SAC. The data distribution is relatively close to the straight line, indicating that the regression model can predict the SAC value of cellulose–silica aerogels.

Figure 2a–f shows the variable interaction of the 3D surface and contour plots on SAC response. The orange–red compositions are desirable, while the green–blue compositions are less desirable. In Figure 2a, the optimum results were obtained by adding 35 wt % cellulose and 15 wt % silica. This combination produces a high level of SAC due to the acoustic energy mostly absorbed by the interface between the cellulose fiber and the silica particles.26 However, adding cellulose and silica content above the optimum point causes a decrease in the SAC value.

In Figure 2c, the reduction in SAC is due to the decreasing cellulose and increasing ZnO concentration. This phenomenon is similar to the results of the studies conducted by Kamal Mohamed et al.63 that stated that adding ZnO increased cellulose solubility and molecules, resulting in a denser mesh-like surface structure. Since it is difficult for sound waves to pass through larger pores in cellulose, less sound is absorbed. Figure 2e shows that increasing silica and decreasing ZnO concentration improve the SAC value. This phenomenon is related to a smaller silica particle size and more uniform distribution, improving acoustic insulation performance.76

The numerical optimization used the desirability function to get the maximum SAC value. Therefore, the optimization aimed to select the cellulose, silica, and ZnO concentration in range and maximize the SAC value. Figure 3 shows the chosen solution formula with the highest desirability level of 1.000, 39.8578 wt % cellulose, 16.5428 wt % silica, 0.5684 wt % ZnO, with an SAC value of 0.9758. After the validation procedure, the measured SAC for the solution formula was 0.9709, with a percentage difference of 0.5%, indicating an outstanding model precision.

Figure 4 shows the SAC value of the cellulose aerogel and cellulose–silica aerogel at different frequencies (50–6400 Hz). The SAC value is directly proportional to the increase in frequency. However, the increase in the sound absorption coefficient was dominant between 2500 and 5000 Hz. Additionally, the cellulose aerogel and cellulose–silica aerogel had the highest SAC values of 0.59 and 0.70 at approximately 4200 and 5000 Hz, respectively. This proves that the incorporation of silica into the cellulose aerogels increased the SAC value, indicating better sound absorption.

X-ray Diffraction (XRD) Analysis. Figure 5 shows the XRD pattern of leached silica, cellulose aerogel, and cellulose–silica aerogel. The round peak appeared at around $2\theta = 22.40^\circ$ for the amorphous silica.35,77 Meanwhile, the sharp crystalline peaks appeared at $2\theta = 12.30, 20.00$, and $21.40^\circ$ for the cellulose aerogel.78,79 Furthermore, these peaks became weaker and appeared at approximately $2\theta = 15–30^\circ$, showing that amorphous silica was formed, and the crystalline structure of the cellulose aerogel was significantly destroyed.80 The explanation for this behavior is probably related to the disruption mechanism, in which the silica particles’ in situ composite on the cellulose surfaces disrupts the bundling of fiber into ribbons.81 These results further show the successful synthesis of cellulose–silica aerogel using waste newspaper and geothermal silica as raw materials by the ambient pressure drying.

Fourier Transform Infrared (FTIR) Spectroscopy. Figure 6 shows the FTIR spectra of leached silica, cellulose aerogel, and cellulose–silica aerogel, while Table 3 summarizes all the FTIR results, which are consistent with previous studies.67,68,82–84 For leached silica, there was no transmittance peak identified between 2800 and 3000 cm$^{-1}$, showing that it is purely used and free from organic matter.85 Furthermore, the peaks for the cellulose–silica aerogel at 3336, 1156, and 1546 cm$^{-1}$ were related to the overlapping effect between silica and cellulose.68 The band at 3336 and 1156 cm$^{-1}$ corresponds to the O–H stretching vibration and Si–O–Si asymmetric stretching vibration, respectively. The results show that silica was impregnated into the cellulose aerogel.

Porous Structure Analysis. Figure 7 shows nitrogen adsorption–desorption isotherms of cellulose aerogel and cellulose–silica aerogel. Type IV isotherms characterize the entire sample, typical of mesoporous materials in the range of 2–50 nm, and are similar to the previous study.64 Table 4 shows that cellulose aerogel and cellulose–silica aerogel have a surface area of 0.60 and 5.63 m$^2$/g, respectively. The greater
surface area of cellulose–silica is due to the silica particles attached to the cellulose fibers, preventing them from shrinking during the regeneration process.\textsuperscript{68} However, the surface area of cellulose–silica aerogel is less than that in the study conducted by Demilecamps et al. (220 m\textsuperscript{2}/g)\textsuperscript{66} due to the collapse of the pores during the process of ambient pressure drying.\textsuperscript{86} Furthermore, the pore size of the cellulose aerogel was 34.19 nm, and that of the cellulose–silica aerogel decreased by 22.00 nm due to the highest silica attached to the fiber surface. In this study, silica was combined with water to become “swollen”, and its particles filled the pore of cellulose and functioned as an “adhesive”, resulting in a decrease in pore size.\textsuperscript{87}

**Scanning Electron Microscope–Energy Dispersive X-ray (SEM–EDX) Analysis.** Figure 8 shows the SEM images of cellulose aerogel and cellulose–silica aerogel with significantly different morphologies. Figure 8a shows long cellulose fibers with a rough surface and relatively large pores, as listed in
Table 4. In contrast, Figure 8b shows silica particles attached and evenly distributed to the surface of the fibers. During the regeneration process, the silanols in the silica sol combine with the cellulose molecules through the dehydrating action between the hydroxyl groups. Conclusively, the silanols covering the surface of cellulose fibers continue to condense, which result in complex structures. In addition, the hydrogen bonding among cellulose, silica, and solvent enhanced the homogeneous network structure, hence, the silica forms finer cellulose fibers.

Figure 9 shows the elemental composition analysis of the cellulose aerogel and cellulose−silica aerogel. The cellulose aerogel contains C, O, Na, Ca, and Zn elements, while the cellulose−silica aerogel contains C, O, Al, Si, Cl, K, Ca, and Zn elements. Furthermore, EDX confirmed the presence of silicon inside the cellulose−silica aerogel, while other elements are probably derived from geothermal silica. The
signal for the silicon atoms and the carbon and oxygen peaks from the cellulose network is identified at 1.74, 0.28, and 0.53 keV, respectively. This result is similar to a previous study. Meanwhile, the EDX analysis is only used to verify the presence of silicon atoms and not to quantify their amount and location.

**Thermal Gravimetric Analysis (TGA).** Figures 10 and 11 show the cellulose aerogel’s and cellulose−silica aerogel’s TG/DTG curves, respectively. The thermal decomposition of cellulose aerogel consists of three phases, namely, 25−105 °C with 10.38%, 106−370 °C with 60.26%, and 371−900 °C with 10.66% weight loss. The evaporation of adsorbed water and some organic components occurs in the first, while the second and third stages are associated with the degradation and decomposition of cellulose and some organic components. Therefore, the mass of most cellulose aerogel is lost at this stage. Furthermore, when heated to a higher degree, the cellulose aerogel loses its structural stability, resulting in a total collapse.

The thermal decomposition of cellulose−silica aerogel consists of four phases, namely, 25−105 °C with 12.77%, 106−350 °C with 39.89%, 351−500 °C with 8.57%, and 501−900 °C with 5.57% weight loss. At temperatures above 500 °C, the residual ash generally consists of silica. The cellulose−silica aerogel has a smaller mass loss compared to the cellulose aerogel. This is due to the increase of silica and the decrease of cellulose content in composite aerogels, which cause the weakening of their synergistic effect and also the transformation of the framework structure from cellulose to silica-dominated. However, the curves showed that the cellulose−silica aerogel has a significant weight loss at 300−400 °C with the presence of ZnO, where lignocellulose was highly sensitive to inorganic salts as reported in a previous study. At 900 °C, the weight loss of cellulose−silica aerogel was 66.80%, indicating that silica can significantly stabilize the cellulose against decomposition.

Table 5 shows the previous studies on SAC from the material-based aerogel. Although the cellulose−silica aerogel was made from recycle cellulose fiber and geothermal silica, it has a high SAC value compared to several previous studies. Sadly, this value is lower than that in the research conducted by Silviana et al. This is probably due to the use of different chemicals to improve the solubility of NaOH solution. The previous study used polyethylene glycol (PEG), which can stabilize cellulose solution. However, ZnO has the ability to form stronger hydrogen bonds with cellulose, which improve the solubility of cellulose. The addition of ZnO to the NaOH/water solution leads to the production of smaller particles and a hydroxide layer (≡Zn−OH) on the particle surface. These hydroxide species readily bind to water and reduce the volume of water molecules close to the cellulose chain, hence inhibiting aggregation and delaying gelation.

Furthermore, the cellulose−silica aerogel belongs to sound absorption class A (ISO 11654), which is the best sound absorption category.

**CONCLUSIONS**

This study successfully synthesized cellulose−silica aerogel using waste newspaper-based cellulose, geothermal silica, and NaOH/ZnO solution with ambient pressure drying. The result
Figure 9. EDX mapping of (a) cellulose aerogel and (b) cellulose–silica aerogel.

Figure 10. TG/DTG curves of cellulose aerogel.

Figure 11. TG/DTG curves of cellulose–silica aerogel.
structure due to the hydrogen bond among cellulose, silica, and sound-insulating material. Furthermore, the distribution of this shows a significant model with improving the thermal stability of cellulose to the pore size of cellulose solvent. Its presence increases the surface area but decreases shows that the silica impregnated cellulose aerogel is a better Figure 12. Scheme of cellulose–silica aerogel preparation.

Table 5. Comparison of SAC Value with Other Studies

| Material                        | Method                   | t (cm) | Frequency (Hz) | SAC   | Author                |
|---------------------------------|--------------------------|--------|----------------|-------|-----------------------|
| MTMS–VTMS derived aerogel       | impedance tube           | 1.6    | 200–2500       | 0.9   | Cai et al.¹⁰          |
| polymide-silica aerogel         | impedance tube           | 1 and 3| 2500–10,000    | 0.8   | Yan et al.¹¹          |
| silica aerogel, GSA-SDS aerogel | impedance tube           | 1–5    | 50–1600        | 0.86  | Sachithanadam and Joshi ⁹² |
| cellulose–silica aerogel        | impedance tube           | 1      | 50–6400        | 0.70  | this study            |
| monolithic silica aerogel       | impedance tube           | 1.27–2.54| 100–4900    | 0.88  | Merli et al.⁹³        |
| silica–cellulose aerogel        | sound level meter        | 1      | 31.5–8000      | 0.39–0.50 | Feng et al.³⁵       |
| silica–cellulose aerogel        | sound level meter        | 1      | 31.5–8000      | 0.9896| Silviana et al.²⁵      |
| cellulose–silica aerogel        | sound level meter        | 1      | 31.5–8000      | 0.9709| this study            |

Figure 12. Scheme of cellulose–silica aerogel preparation.

shows that the silica impregnated cellulose aerogel is a better sound-insulating material. Furthermore, the distribution of this compound on the surface of cellulose enhances the network structure due to the hydrogen bond among cellulose, silica, and solvent. Its presence increases the surface area but decreases the pore size of cellulose–silica aerogel. Silica also contributes to improving the thermal stability of cellulose–silica aerogel. Additionally, the optimum SAC was obtained at 39.8578 wt % cellulose, 16.5428 wt % silica, 0.5684 wt % ZnO. The results of ANOVA show a significant model with p value, R², and adj R² of 0.0252, 97.74%, and 90.94%, respectively. These results are essential for the future development of cellulose–silica aerogels, especially for the improvement of drying methods.

### MATERIALS AND METHODS

**Materials.** The waste newspaper was collected locally in Semarang, and geothermal silica was supplied from PT Geo Dipa Energi, Dieng, Indonesia. Furthermore, zinc oxide (ZnO) powder was purchased from SmartLab in the same country. Sulfuric acid (H₂SO₄, purity 98%), sodium hydroxide (NaOH, purity 98%), and citric acid (C₆H₈O₇, purity 98%) were purchased from Merck, Germany, and used without further purification. Finally, the preparation was performed using distilled water.

**Methods.** Pretreatment of Geothermal Silica. Pretreatment was performed to remove organic and residual impurities in geothermal silica based on the procedures in previous studies but with some modifications.⁹⁵,⁹⁶ Geothermal silica was dried at 105 °C for 24 h and mixed with sulfuric acid at 90 °C for 3 h, and the ratio of silica to H₂SO₄ was 1:5 (w/v). After the slurry was formed, the solution was screened through a filter paper to separate sodium silicate (Na₂SiO₃) and the solid residue.

**Preparation of Cellulose from Waste Newspaper.** The waste newspaper was treated by chemical extraction to remove ink and glue based on the procedures in the previous studies.¹⁷,¹⁸ It was cut into small pieces (1–2 cm), soaked in a 2 N sodium hydroxide solution, heated at 80 °C, and stirred for 3 h. Afterward, it was left overnight at ambient temperature. The mixture was then screened and washed with distilled water at least five times until a neutral pH was obtained.

**Preparation of Cellulose Aerogel.** The method used for the cellulose aerogel preparation is based on the procedures in the previous studies with some modifications.⁵² Cellulose and NaOH/ZnO aqueous solution were stirred for 1 h, frozen for 12 h at −15 °C, and then thawed at ambient temperature with constant stirring for 30 min. The cake was immersed in 0.2 M citric acid to neutralize sodium hydroxide after being frozen at −15 °C for a further 12 h. The solution was filtered and washed with distilled water until it reached a neutral pH. Subsequently, it was dried in an oven at 90, 100, and 110 °C for 90 min each.

**Preparation of Cellulose–Silica Aerogel.** The preparation of cellulose–silica aerogel as shown in Figure 12 is based on the procedures in the previous studies with some modifications.⁶²,⁶⁶ First, cellulose, Na₂SiO₃, and NaOH/ZnO aqueous solution were stirred for 1 h. Furthermore, the solutions were frozen for 12 h at −15 °C and then thawed at ambient temperature with constant stirring for 30 min. The cake was immersed in 0.2 M citric acid to neutralize sodium hydroxide after being frozen at −15 °C for a further 12 h. The solution was filtered and washed with distilled water until it reached a neutral pH. It was then dried in an oven at 90, 100, and 110 °C for 90 min in each.

**Design Experiment.** The Box–Behnken design using the Design-Expert software (version 12.0) was used to determine the effect of cellulose, silica, and ZnO concentrations on SAC value. Table 6 shows the coded levels for these variables. The
ZnO variable was based on the previous study of Demilecamps et al., using an upper and lower range approach. However, the cellulose, silica, and ZnO were mixed with varying concentrations and dried for different periods in a preliminary study.

**Measurement of Sound Absorption Coefficient.** The methods used to measure the SAC of cellulose—silica aerogel in this study are a sound level meter and an impedance tube.

**Study of the Sound Level Meter.** This method was conducted according to a previous study. Figure 13 shows that the sound absorption coefficient of the cellulose—silica aerogels with a thickness of 10 mm was studied using a sound level meter (Lutron SL-4011) and a sound signal generator (Blesi Guardian Angel self-protection alarm, 90 dB). The sound generator was housed inside an insulation box made by fixing cellulose—silica aerogels at all sides to ensure a closed system. Furthermore, the incident sound signal was measured 400 mm away from the generator source.

The following equation is used to calculate the sound absorption coefficient measured by the sound level meter:

\[
Li = 10 \log \left( \frac{I}{I_0} \right) \text{dB}
\]

where \( Li \), \( I \), and \( I_0 \) are the sound intensity level (dB), the sound intensity \((\text{W/m}^2)\), and the reference sound intensity \((10^{-12} \text{ W/m}^2)\).

The sound absorption coefficient (SAC) is the fraction of sound energy absorbed by the material. It represents a value in the range of 0 to 1. It can be expressed as:

\[
\alpha = 1 - \frac{I_0}{I} \tag{3}
\]

where \( \alpha \), \( I_0 \), and \( I \) are the sound absorption coefficient, the reflected sound intensity, and the incident sound intensity.

**Study of the Impedance Tube.** An impedance tube compared the sound absorption coefficient measured using the sound level meter (the theoretically calculated value). This is because it is a compact setup that can easily and affordably determine small samples’ absorption data. Furthermore, the optimum SAC value was used to measure cellulose—silica aerogel and cellulose aerogel (silica free) with a 3 cm diameter and a 1 cm thickness. Figure 14 shows that the Bruel & Kjær Impedance Tube Kit Type 4206 measured the sound absorption coefficient at a frequency in the range of 50 Hz to 6.4 kHz. Additionally, the absorption coefficient measurements using the two-microphone transfer-function method were according to ISO 10534-2 and ASTM E1050-12 international standards. The measurements were made with two \( 1/4 \) in Condenser Microphones Type 4187, fitted with Type 4206, specially designed to reduce errors due to pressure leakage at high frequencies. Meanwhile, the sound absorption coefficient was calculated from the sound pressures in the incident and reflected wave.

**Characterization.** The structures of the aerogels were analyzed using an X-Ray diffractometer (XRD, PANalytical X’Pert PRO). The patterns of XRD were recorded in the 2θ range of 10 to 90°. Furthermore, their functional groups were identified using an FTIR spectrophotometer (Perkin-Elmer UATR Two) with the ATR method. The IR spectra were recorded from 4000 to 450 cm\(^{-1}\). The morphology of aerogels was investigated using a scanning electron microscope (SEM, Jeol JSM-IT200) at an acceleration voltage (VACC) of 5 kV. Also, the distribution and elemental composition of silica particles were determined using an energy-dispersive X-ray (EDX) spectrometer, which was coupled with the SEM. The nitrogen gas sorption/physi sorption was measured at \(-195.85 \text{°C} \) using Micromeritics Tri Sar II 3020 Version 2.00. Additionally, the surface area of aerogels and the pore size distributions were determined using Brunauer—Emmett—Teller (BET) and Barrett—Joyner—Halenda (BJH) methods, respectively. Finally, their thermal stability was analyzed using thermogravimetric analysis (TG) with the simultaneous thermogravimetric analyzer NEXTA STA (Hitachi STA200RV with real view sample observation). The samples were heated in flowing air from ambient temperature to 900 °C at 10 °C min\(^{-1}\).

**ECONOMIC ANALYSIS OF AEROGEL PRODUCTION**

The economic analysis of aerogel production should be as detailed as possible, with all the important elements included. This study is focused on the material and energy costs, which are the most crucial parameters to consider. Table 7 shows the material costs that we used. Since the cellulose fiber was extracted from waste newspaper and silica from geothermal solid waste, these materials are available free of charge. However, transportation costs for silica are $40, and transportation costs for other materials are the responsibility of the company that supplies raw materials for aerogel manufacturing.

The following equation expresses the energy costs required to manufacture 1 kg of aerogel:

\[
\text{Energy Costs} = \frac{\text{Costs of Materials} + \text{Transportation Costs}}{\text{Production Value}}
\]

**Table 6. Coded Level BBD**

| variable         | symbol | coded level |
|------------------|--------|-------------|
| cellulose (wt %) | \( x_1 \) | 30 35 40    |
| silica (wt %)    | \( x_2 \) | 10 15 20    |
| ZnO (wt %)       | \( x_3 \) | 0.5 1 1.5   |

**Figure 13. Measurement of SAC using the sound level meter.**

**Figure 14. Measurement of SAC using the impedance tube.**
Garrido et al. studied the manufacture of inorganic aerogel (IA) and two hybrid aerogels produced in powder (HYB-A) and monolithic (HYB-C) forms. Table 9 and Figure 15 show an overview of the total material cost for several types of aerogels. When compared to the silica-based aerogels, cellulose–silica aerogel (CSA) is the cheapest acoustic insulating material despite the fact that there is a $40 transportation cost. It is obvious that the utilization of recycled materials reduces the production cost of cellulose–silica aerogel and can be scaled up for industrial applications.

Table 7. Material Costs for the Synthesis of Cellulose–Silica Aerogel

| material       | cost ($/kg of aerogel) |
|----------------|------------------------|
| ZnO            | 0.8                    |
| H₂SO₄ (98%)    | 50                     |
| NaOH (98%)     | 40                     |
| C₆H₈O₆ (0.2 M) | 60                     |
| distilled H₂O  | 0.9                    |

\[ C = T \times n \times P \]  \hspace{1cm} (4)

where \( C \), \( T \), \( n \), and \( P \) are the energy cost ($/kg of aerogel), 1 kWh of electricity cost ($/kWh), number of operating hours (h), and power (W). The energy costs are calculated according to eq 4, as shown in Table 8.

Table 8. Energy Costs for the Synthesis of Cellulose–Silica Aerogel

| process      | energy cost ($/kg of aerogel) |
|--------------|--------------------------------|
| stirring     | 0.01                           |
| freezing     | 0.24                           |
| drying       | 0.56                           |

Table 9. Total Cost of the Aerogels

| aerogel | cost ($/kg of aerogel) | material | energy | transport | total costs |
|---------|------------------------|----------|--------|-----------|-------------|
| CSA     | 151.7                  | 0.81     | 40     | 192.51    |
| IA      | 439.83                 | 12.66    |        | 588.66    |
| HYB-A   | 645.03                 | 3.19     |        | 648.22    |
| HYB-C   | 722.03                 | 12.66    |        | 734.69    |

Figure 15. Total cost for aerogel production.

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**Notes**

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**ABBREVIATIONS**

ZnO, zinc oxide; NaOH, sodium hydroxide; BBD, Box–Behnken design; SAC, sound absorption coefficient; XRD, X-ray diffraction; FTIR, Fourier-transform infrared; SEM–EDX, scanning electron microscopy—energy-dispersive X-ray; BET–BJH, Brunauer, Emmett, Teller–Barrett, Joyner, Halenda; TGA, thermal gravimetric analysis

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