# FLY ASH-BASED AEROGEL REINFORCED WITH POLYESTER FIBRES FOR REMARKABLE THERMAL AND ACOUSTIC PROPERTIES

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ABSTRACT: A nonconventional material with remarkable thermal and acoustic insulation properties from aerogels category was experimentally made. Produced from a recycled commercial waste (polyethylene terephthalate-PET) and an industrial by-product (fly ash) as well as a biomaterial (psyllium husk) used for the first time as a binder by mixing with water, the aerogel was subjected to a freeze-drying process. The determination of the product characteristics highlighted extremely low densities (0.022-0.041 g·cm$^{-3}$), very low thermal conductivities (0.035-0.037 W·m$^{-1}$·K$^{-1}$), very high porosities (97.8-98.5 %), mechanical properties superior to those of an unreinforced gel (compression strength of 9.5-16.4 kPa and Young's modulus of 4.15-18.93 kPa), and excellent noise absorption properties (noise reduction coefficient of 0.30-0.33). The manufacturing process of this inexpensive product allows for a clean environment.
KEYWORDS: aerogel, fibre, insulation properties, acoustic, fly ash, psyllium husk.

## 1. INTRODUCTION

The first information regarding the design of an aerogel by extracting the liquid component of the traditional gel and its replacement with a gas in order to significantly reduce the density (below 0.5 g·cm$^{-3}$), and thermal conductivity (in the range 0.012-0.050 W·m$^{-1}$·K$^{-1}$) as well as the increase of porosity (over 95 %) is from the middle of the last century [1]. Subsequent research in this field was generally developed in the 3$^{rd}$ millennium [2] and the types of aerogel were much diversified (using a wide variety of materials) from silica aerogel [3], to carbon-based aerogel [4], cellulose-based aerogel [5], composite aerogel [6], and metal aerogel [7]. The remarkable properties of aerogels make possible the existence of numerous areas of their application: thermal insulation, acoustic insulation, chemical adsorption, catalyst support, solar radiation and thermal emittance, electromagnetic shielding, components in energy absorption, cosmetics, cosmic dust collection (only silica aerogel), etc. [8].

Recent concerns of researchers in the world are focused on the manufacture of composite aerogels using waste or industrial by-products that affect the quality of environment by their storing in landfills. According to the paper [9], the rubber fibres from degraded tires were introduced in the rubber aerogel production process using the freeze-drying process. The aerogel manufactured by this technology had a very porous structure (porosity over 90 %) with an extremely low density (between 0.02-0.091 g·cm$^{-3}$). From a mechanical point of view, the material was robust, with the Young's modulus reaching 965.6 kPa. The thermal conductivity had extremely low values (0.035-0.049 W·m$^{-1}$·K$^{-1}$) and the sound absorption was very efficient (the noise reduction coefficient dropping to 0.56). The research presented in this paper showed valuable results for the use of rubber aerogel in applications aimed at the thermal and acoustic insulation of buildings, automobiles, and aircraft.

Other available residual materials suitable for aerogel manufacturing were experimental tested in the work [10]. Fly ash as an industrial by-product of energy industry resulting from burning the coal was mixed with recycled polyethylene terephthalate fibre for reinforcing the final product, the binding of the two ground waste being facilitated by the use of xanthan gum aqueous solution with the concentration between 0.5-3 %. The gel poured into a tray was then lyophilized (freeze-drying) forming a highly porous structure. The resulting composite aerogel had remarkable properties (very low density in the range of 0.026-0.062 g·cm$^{-3}$, low thermal conductivity between 0.034-0.039 W·m$^{-1}$·K$^{-1}$, high porosity in the range of 96.6-98.4 %, variable Young's modulus of 4-20.6 kPa, and coefficient of noise reduction within the limits 0.18-0.31. The experiment offered an ecological and cheap technical solution for recycling the fly ash turned into valuable materials.

The paper [11] describes an aerogel manufacturing process that also uses coal fly ash provided by the energy industry. Unlike the process shown in [10], polyvinyl alcohol (99 %) in the form of flakes was used for reinforcing. The mixture of the two types of raw material was made in the following weight proportions: 2.5 % fly ash, 2 % polyvinyl alcohol, and 100 mL of deionized water. Methyltrimethoxysilane (95 %) was added as a binder. The gel was poured into a tray. The freeze-drying process took place in two stages: first in a freezer at -10 ºC for 12 hours, and the second in a lyophilizer at -50 ºC for 24 hours. The role of applying the freeze-drying method was the minimization of the surface tension effect leading to a highly porous structure without affecting it. The fly ash aerogel made by the described above method had low density of 0.10-0.19 $g \cdot cm^{-3}$, high porosity reaching 90 %, low thermal conductivity in the range of 0.042-0.050 $W \cdot m^{-1} \cdot K^{-1}$, and noise reduction coefficient of 0.20-0.30. The Young's modulus reached the maximum value of 150 kPa.

The current work aimed at the manufacture of a high-performance aerogel under the conditions of using the available reserves of residual materials (coal fly ash and recycled polyethylene terephthalate bottle for its fibres). The work originality compared to the solutions presented above and in particular, compared to the manufacturing recipe applied in [10], was the nature of the binder adopted in the experiment based on psyllium husk powder, a soluble fibre. Its binding ability is activated by mixing this powder with water. Psyllium husk is obtained from Plantago Ovata, a plant cultivated in several countries (e.g. Spain, France, India). It is commercially available as a pharmaceutical product. The preparation of this gel consists in dissolving by mixing 5-20 g of psyllium husk powder in 100 g of deionized water. The obtained aqueous solution plays the role of binder. According to the basic principle of aerogel manufacturing, the freeze-drying technique of the semi-prepared gel was also used in this work.

## 2. METHODS AND MATERIALS

### 2.1 Methods

The main characteristic of the manufacturing technique of an aerogel with excellent performance regarding the extremely low thermal and acoustic insulation of the product is achieving the freeze-drying process at around -50 ºC of the gel obtained by mixing and dissolving the solid components in the aqueous solution of the binder. The significant decrease of the surface tension effect in the gel mass is the major purpose of applying the freeze-drying method, avoiding the destruction of the material structure due to the excessive increase of its porosity.

On the other hand, the structural reinforcement of aerogel is pursued through the use of raw materials that include fibres, as it is known that they substantially contribute to the improvement of mechanical properties of the composite [12-14]. The experiment described in this paper adopted the use of finely chopped recycled PET (polyethylene terephthalate) bottles as a fibre supplier.

The component operations of the aerogel manufacturing method occurred in the following chronological order. The preparation of solid materials including fly ash and PET glass consisted of fine grinding (below 20 μm) of the ash supplied by the Paroșeni thermal power plant (Romania) and small chopping into pieces below 2 mm of PET glass collected by the authors. The two raw materials were mixed for homogenization with a mixer. The binder was prepared by dissolving and mixing 5-20 g of psyllium husk powder in 100 g of deionized water. Then, the binder was poured over the solid homogenized mixture deposited in the 100 W-VEVOR electric stirrer mixer. The mixing operation was performed at 250 rpm for 30 min. Next, the obtained gel was poured into a tray and then it was inserted into the BIOBASE BOV-T140C device with the capacity of 137 L, in which the freeze-drying process took place at -50 C. The duration of the process was divided into two rounds of every 8 hours.

### 2.2 Materials

The materials used during the experiment were: coal fly ash whose oxide composition included 46.5 % $SiO_2$, 7.9 % CaO, 3.2 % MgO, 10.1 % $Na_2O$ and $K_2O$, 23.7 % $Al_2O_3$, 8.6 % $Fe_2O_3$, recycled PET bottle that is a polymer produced by polymerization of ethylene glycol and terephthalic acid, psyllium husk powder composed of arabinose (22 %), xylose (67 %), uric acids (10-15 %) with low amounts of galactose, rhamnose, glucose and mannose [15], and deionized water.

### 2.3 Characterization methods of aerogel samples

Generally, the methods for characterizing the aerogel samples made by the technique presented above were selected from those frequently used. Having regular geometric shapes, the sample density was determined as the ratio between its mass and volume [16]. The aerogel porosity was measured by the saturation or inhibition method presented in the work [17]. Thermal conductivity was determined using a Heat-Flow Meter HFM 446 Lambda (according to SR EN 1946-3:2004), from NETZSCH at room temperature. 10 kN-Hydraulic Uniaxial Press Machine was used according to EN 826: 2013 for measuring the compression strength of samples [18]. Compression and decompression cycles of the samples were made until their deformation of 25 % was reached (with the rate of 1 mm/min), thus indicating the value of Young's modulus and also the recovery proportion of the specimens subjected to the test. The method adopted for determining the sound absorbing performances of the aerogel was that presented in the paper [19]. The measurement of the sound absorption coefficient was performed in a standing wave tube, vertically placed, having a speaker at one end, while the sample subjected to the test was placed at the opposite end. Microstructural images (SEM) of aerogels were obtained with Biological Microscope MT5000 model with captured image, 1000 x magnification.

## 3. RESULTS AND DISCUSSION

### 3.1 Results

As mentioned above, three experimental variants were chosen to produce aerogel based on residual materials (fly ash and PET bottle) aiming to obtain excellent thermal and acoustic insulation performances.

Table 1 contains the main data on the weight proportions of the components of aerogel composite.

**Table 1.** Composition of experimental variants

| Variant | Raw material (wt. %) | | Binder (g) | |
|---|---|---|---|---|
| | PET bottle | Fly ash | Psyllium husk | Deionized water |
| 1 | 2 | 1.0 | 5 | 100 |
| 2 | 2 | 2.2 | 12 | 100 |
| 3 | 2 | 3.5 | 20 | 100 |

According to Table 1, the fibre supplier with the role of reinforcing the composite (i.e. PET bottle) was kept in a constant proportion of 2 wt. %. Instead, the amount of raw material that contributes to the decrease of aerogel density and increase of its porosity (i.e. fly ash) was increased from 1 wt. % up to 3.5 wt. %. Also, the binder adopted in this experiment (psyllium husk), which when mixed with water significantly activates its binding ability, was used in increasing quantities from 5 g to 20 g.

Images of aspect of reinforced aerogel specimens are shown in Figure 1.

The characteristics of the aerogel composite in the three variants determined by the methods mentioned above are centralized in Table 2.

**Table 2.** Characteristics of the aerogel composite

| Variant | Density ($g \cdot cm^{-3}$) | Porosity (%) | Thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$) | Compression strength (kPa) | Young's modulus (kPa) | Noise reduction coefficient (at 1300 Hz) | Aerogel composite | |
|---|---|---|---|---|---|---|---|---|
| | | | | | | | Volume ($cm^{-3}$) | Mass (g) |
| 1 | 0.041 | 97.8 | 0.037 | 9.5 | 4.15 | 0.30 | 180 | 7.38 |
| 2 | 0.031 | 98.2 | 0.036 | 12.9 | 11.34 | 0.31 | 180 | 5.58 |
| 3 | 0.022 | 98.5 | 0.035 | 16.4 | 18.93 | 0.33 | 180 | 3.96 |

a b c

**Figure 1.** Aspect of reinforced aerogel specimens
a – variant 1; b – variant 2; c – variant 3.

The experimental results presented in Table 2 confirm the remarkable thermal and acoustic insulating properties of the aerogel prepared with PET bottle (containing fibres for reinforcing) and fly ash as raw materials as well as a vegetable binder (psyllium husk) with binding ability activated by mixing with water. The freeze-drying method adopted worldwide led to obtaining the density and thermal conductivity at extremely low levels (between 0.022-0.041 $g \cdot cm^{-3}$ and respectively, 0.035-0.037 $W \cdot m^{-1} \cdot K^{-1}$). The very high porosity of the aerogel (97.8-98.5 %) decisively contributed to the significant increase of the noise reduction coefficient (between 0.30-0.33 corresponding to the noise frequency below 1300 Hz). Due to the reinforcement of the aerogel with fibres through the contribution of PET bottle from the composition of the starting mixture, the mechanical properties of an extremely light material such as aerogel were significantly increased compared to an unreinforced aerogel (compression strength reached 16.4 kPa and Young's modulus recorded the maximum value of 18.93 kPa). It was experimentally observed that under the conditions of keeping constant the amount of PET bottle, increasing the proportion of fly ash in the starting mixture composition contributed to the decrease of density and thermal conductivity of the aerogel, while the porosity and the noise reduction coefficient had higher values. During the freeze-drying process, the mass loss was almost twice as high in the variant using the maximum proportion of fly ash (variant 3) under the conditions where the specimens volume was constant (about 180 $cm^{-3}$) in all the tested variants.

Figure 2 presents microstructural images of reinforced aerogel specimens Common characteristic of these images is the presence of reinforcing fibres. The picture representing variant 3 has a more robust distribution of fibres contributing to the improvement of its mechanical properties.

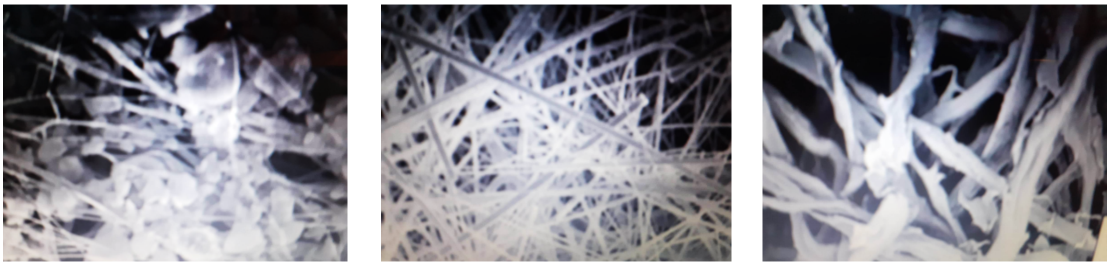


**Figure 2.** Microstructural configuration of reinforced aerogel specimens: a—variant 1; b—variant 2; c—variant 3.

3.2 Discussion

The most important stage of aerogel manufacturing is freeze-drying. This process has the advantage of keeping the porous structure of the gel unchanged during its development. The presence of fibres in the aerogel matrix ensures the necessary resistance against the lateral capillarity tension exhibited during the drying process. It was experimentally confirmed that the fibres act as supporting skeletons avoiding the destruction of the porous structure as a result of the excessive increase of the gel porosity.

The originality of the current work consists in the choice for the first time of a binder based on a vegetable material (psyllium husk) commercially available as a pharmaceutical product. Mixing it with deionized water activates its excellent binding ability.

The aerogel presented in this paper is part of the current concerns of the world research to create high-performance products based on materials available as waste or industrial by-product. Except for the binder mentioned as a biomaterial, the aerogel manufacturing process included an industrial by-product (coal fly ash) and a commercial waste (recycled PET bottle). The use of these residual materials allows keeping a clean environment and the manufacturing process is cheap. The properties of the new aerogel designed and tested in this work were almost similar to those previously manufactured.

## 4. CONCLUSION

The production of an aerogel with remarkable thermal and acoustic insulation properties using cheap residual materials (fly ash, PET bottle) and a biomaterial (Psyllium husk) for the binder preparation was the objective of the current work.

The work originality is the choice for the first time of a binder based on a biomaterial (Psyllium husk).

The properties of the new aerogel (extremely low density and thermal conductivity, very high porosity, excellent noise absorption properties, acceptable compression strength, and Young’s modulus) were almost similar to those previously manufactured in the world.

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