Development of advanced aerogel-based composite material with high performance for building industry

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Abstract
According to the European Commission's plan, greenhouse gas emissions in the European Union must be reduced by 80% compared to the level from year 1990 (see [1]). In order to reduce the energy consumption of the buildings, an optimization of the building insulation is an effective measure. Super-insulating materials are promising materials to fulfil these objectives. Present work describes development of advanced aerogel-based composite material with small thickness. Such composite materials based on silica aerogel can be extremely efficient with regard to their thermal insulation properties. In this study, the experimental investigations of hydrothermal and mechanical performance were conducted on the aerogel-based insulation blanket and its constituents (core material and aerogel granules). Furthermore, the effect of ageing to performance of such material is assessed. The developed aerogel-based insulation material is characterized by very low thermal conductivity (under 18.0 mW/(m·K)) and good hydrothermal properties. It has been shown here that the thermal conductivity of an insulating material made of glass fibre can be reduced to more than half with using of aerogel granules. This aerogel-based composite material is characterized by good hydric properties. The material is both hydrophobic and water vapor permeable. In addition, mechanical properties of new composite material fulfill the multifunctional application of this promising insulating material. New product can be used for external thermal insulation system (ETICS) as well as for internal thermal insulation system. Copyright © 2018 VBRI Press.

Keywords: aerogel, insulation, composite material, hydrothermal, mechanical, ageing.

Introduction
According to the European Commission's plan, greenhouse gas emissions in the European Union must be reduced by 80% compared to the level from year 1990 (see [1]). In order to reduce the energy consumption of the buildings, an optimization of the building insulation is an effective measure. A traditional solution would be insulation with thicker products. This option is associated with higher costs, reduction of usable area and negative environmental aspects.

Within the scope of the research project "Homeskin" (see [2]) a high performance insulating material with small thickness is developed. Since the traditional insulating materials (e.g. mineral wool, extruded polystyrene foam, expanded polystyrene) have generally reached their optimization limits, nowadays composite materials are usually developed. This means that conventional insulation materials are usually combined with a suitable nanoporous material in order to further reduce thermal conductivity. In present work, the aerogel-based insulation board was developed and assessed. First studies on the novel aerogel-based insulation materials are made from e.g. [3], [4], [5], [6], [7].

However, the composite products presented in this work were produced in new developed production process. In scope of this innovative manufacturing process, a complete production processes from aerogel until end product is made in one reactor. On that way a supercritical drying of the gel is not included in production chain and use of end product is not limited due to high (production) costs as it is case for the aerogel composite materials available at market.

In the present paper, the experimental investigations on an aerogel-based insulation material are presented. Such composite materials based on aerogel can be extremely efficient with regard to their thermal insulation properties. The thermal conductivity can be between 15 and 20mW/(m·K). The experimental investigations of hydrothermal and mechanical performance were performed on the composite material and its constituents (core material and aerogel granules).
Materials
Aerogel-based insulation boards and its components were tested in scope of this work. In production of this aerogel composite material an innovative ambient drying technique was applied instead of generally used and energy-intensive supercritical drying. The aerogel-based insulation boards are composed of fibrous network (glass fiber) which is impregnated by pre-hydrolyzed Tetraethoxysilane precursors. This gelation process results in a silica organogel which is hydrophobized in next production step. Such composite material is as following dried by fixed frequency electromagnetic waves. In this final stage the fibers take form of board.

Fig. 1 shows SEM images of glass fiber core material and aerogel-based glass fiber board. By comparing to flexible glass fiber core material, the developed aerogel-based glass fiber board is rigid material. The thickness of the aerogel-based material is 20 mm.

The aerogel granulate is a nanoporous material with very good thermal insulation properties. In addition, this material is temperature resistant, pressure resistant and very light (pores up to 99.9 vol.-%). For the developed composite material, a silica aerogel granules were used.

Methodology and material characterization
Thermal properties
Thermal conductivity was measured accordingly to the standard [8], whereby the specimen size of L·B=200·200 mm was chosen. Two-plates apparatus with guarded hot plate was used for measurement. If not mentioned differently, samples were preconditioned at 23 °C and 50% relative humidity prior to testing. As core material is soft, spacers have been used to define thickness of sample. Moreover, aerogel granules were filled to 50 mm high purpose-built frame for measurement of thermal conductivity of loose material. In this way, the bulk density of loose sample could be varied depending on level of compaction of material in frame.

First tests for aerogel granules were performed with standard composition of aerogel without previous sieving. The granules were compacted from bulk density 72.9 kg/m³ up to maximum possible density of 90.5 kg/m³. Furthermore, thermal conductivity was measured for sieved aerogel granules with particle size of 1.0-2.0 and 2.0-4.0 mm. The thermal conductivity (see Fig. 2) varied between 15.9 and 20.7 mW/(m·K), with a significant dependence on the bulk density of the sample. Bulk density of the sieved granules was approximately 65 kg/m³ for both particle sizes 1-2 and 2-4 mm. In these cases, thermal conductivity from approximately 20 mW/(m·K) was reached. Through compaction of particles 2-4 mm to its maximum bulk density, there is minor reduction (1 mW/(m·K)) of thermal conductivity. On other hand, this value correlates with thermal conductivity which would be reached for non-sieved aerogel with same bulk density. The measurements of aerogel show a linear relationship between thermal conductivity and density independent of particle size. These results confirm the investigations of [9] and [10].

Measured thermal conductivity of core material (glass wool) was 33.7 mW/(m·K). This value lies in the lower range of thermal conductivity for mineral wool insulation. For this reason, the material is considered as the good candidate for core material of the aerogel-based insulation board.

Fig. 1. SEM images of (a) core material, (b) aerogel-based material.

Fig. 1. Thermal conductivity of aerogel granules.
In order to measure influence of moisture content to thermal conductivity for aerogel-based insulation board, tests were performed for three different kinds of conditioning, namely 23°C / 50% RH, 70°C / 0% RH (dry) and higher relative humidity (23°C / 80% RH). Samples were conditioned by these climates until saturation prior to testing. Results are shown in Fig. 3. For developed aerogel-based blanket a minor influence of moisture content on thermal conductivity can be seen. Measured thermal conductivity for standard conditioning, 23°C / 50% RH, is 17.2 mW/(m·K). This value lies within the scope of known range for aerogel-based insulation materials available on market, namely between 13.1 mW/(m·K) for Spaceloft [11] from Aspen Aerogels and 19.0 mW/(m·K) for Aerorock ID [12] from Rockwool.

Hydric properties

Main focus of this research lies on hydric behavior of an aerogel-based insulating material, which affects its technical usability in a humid environment and serviceability (condensation issues and mold).

In order to measure moisture absorption and desorption performance of developed aerogel-based boards, the samples were stored at the target relative humidity. Before measurement, samples were dried at the temperature of 70°C. Afterwards the samples were stored at 50% RH, 80% RH, 90% RH and 95% RH until constant mass was achieved. The moisture content is calculated using the following equation:

\[
\Delta m \% = \frac{m_{\text{after}} - m_{\text{initial}}}{m_{\text{initial}}} \times 100 \tag{1}
\]

where \(m_{\text{after}}\) is the mass of the sample at a certain relative humidity and \(m_{\text{initial}}\) is the mass of the dry sample. The sorption isotherm is showed in Fig. 4. After storing the samples at 95% RH, moisture content of 1.6% was observed. This measurement shows low moisture absorption performance of developed aerogel-based insulation and proves hydrophobic characteristics of material.

Water absorption was tested acc. to [13] (short term water absorption) and [14] (long term water absorption). The specimens with size 200 mm \(\times\) 200 mm were conditioned at 23 °C and 50% relative humidity before test. Samples were partially (10 mm) immersed in water. The surface-related water absorption was determined after 24 hours (short term water absorption) and 28 days (long term water absorption). The measured water absorption was 0.14 kg/m² after 24 hours and 0.27 kg/m² after 28 days. Both values fulfill provisions (\(W \leq 1\) kg/m² for short term water absorption and \(W \leq 3\) kg/m² for long term water absorption) given by [15] for mineral wool. Moreover, the water absorption during long-term immersion was clearly below the maximum value (3 kg/m²).

In order to describe liquid transport in the developed aerogel-based material, the water absorption coefficient by partial immersion was measured. This parameter is mostly relevant in case of rain infiltration or high capillarity condensation. The measurement was performed acc. to [16]. The specimens were conditioned at 23 °C and 50% relative humidity before test. Contact with water was possible only with bottom surface of sample. The water absorption coefficient \(A\) was determined as follows:

\[
A \left[\text{kg/(m}^2\cdot\text{h}^{0.5}\right] = \Delta m \ast \sqrt{t} \tag{2}
\]

where \(\Delta m\) is the cumulative water absorption in kg/m² and \(t\) the time in hours. The measured water absorption coefficient by partial immersion is 0.042 kg/(m²·h⁰.⁵). This value is determined for water absorption after 24 hours and shows that tested aerogel blanket is water-resistant material. This measured value is comparable with water absorption coefficient for aerogel-based product for Spaceloft [11] (0.025 kg/(m²·h⁰.⁵)) from Aspen Aerogels.

Water vapor permeability behaviour of the aerogel-based insulation is defined by water vapor diffusion resistance factor \(\mu\). This factor represents the relative magnitude of the water vapor resistance of the material compared to an equally thick layer of stationary air. Water vapor permeability was determined accordingly to [17] and [18]. The samples were preconditioned at

![Fig. 3. Thermal conductivity of developed aerogel-based board for different moisture contents.](image1)

![Fig. 4. Sorption isotherm of developed aerogel-based board.](image2)
23°C and 50% relative humidity prior to testing. Both methods (“dry-cup-method”, for the humidity range from 0 to 50 RH%, and “wet-cup-method”, for the range from 50 to about 100 RH%) were performed because barely any test result for water vapor permeability of aerogel-based boards can be found in the literature. The water vapor diffusion resistance factor μ is determined using the following equation:

\[
\mu[-] = \frac{\delta_{\text{air}}}{\delta} = \frac{\delta_{\text{air}}}{\delta} \cdot \frac{S \cdot \Delta P}{DG \cdot d} \tag{3}
\]

where \(\delta_{\text{air}}\) is the water permeability of the air in the test room condition (\(\delta_{\text{air}} = 0.72 \text{ mg/h}\)), \(\delta\) is the water permeability of the specimen, \(S\) is the exposed area of the test sample, \(d\) is the thickness of the sample, \(\Delta G\) is the mass change of the sample in milligrams per hour after reaching the steady state and \(\Delta P\) is the water vapor pressure difference. Test results show worse material performance for “dry-cup-method” (\(\mu = 5.8\)) than for “wet-cup-method” (\(\mu = 4.5\)). Therefore “dry-cup-method” must be taken for assessment of tested aerogel blanket. Developed material shows expected behavior of water vapor permeability relating to the common insulation materials – mineral wool (\(\mu = 1\)), wood fibre insulation (\(\mu = 1 - 3\)), polystyrene, extruded polystyrene and polyurethane foam (\(\mu = 60 - 150\)). Some materials when exposed to higher temperature and humidity do not keep dimensional stability. This characteristic was tested for developed insulation acc. to [19]. Samples were preconditioned at 23°C and 50% relative humidity prior to testing. Afterwards, the specimens were stored at 70°C and 90% relative humidity. The extension or contraction is calculated from the difference in the dimensions at two target levels. Change of length (\(\Delta L_{\text{L}} = 0.04\%\)) and width (\(\Delta L_{\text{W}} = 0.10\%\)) fulfill requirements given by [15] for mineral wool (\(\Delta L_{\text{L}}, \Delta L_{\text{W}}, \Delta L_{\text{I}} \leq 1\%\)). The dimensional stability should be improved for thickness (\(\Delta L_{\text{I}} = 1.09\%\)).

**Mechanical properties**

Based on experience, it is known that improving of material thermal properties can lead to deterioration of mechanical performance. For better assessment of developed material, tests on mechanical properties were performed. All samples were conditioned at 23°C and 50% relative humidity prior to testing. The results are shown in table 1 and assessed acc. to [15] for mineral wool. Moreover, results are compared to requirements given by [20] for External Thermal Insulation Composite System (ETICS). Tensile strength perpendicular to faces and shear strength comply with mineral wool products with comparable compressive stress by 10% deformation and which are used in ETICS. Because of relatively low tensile strength, the aerogel blankets must be additionally to gluing also anchored to wall. This material can be also used for acoustic insulation for partition walls because of its low compressibility (\(c \leq 2\ mm\)). Although its resistance to point load is relatively high (\(F_p = 587\ N\)), material cannot be used as external roof insulation. Reason for this is that such external roof insulations must have higher compressive stress than here presented material, typically higher than \(\sigma_{10} \geq 60kPa\).

**Table 1. Mechanical properties of developed aerogel-based board.**

<table>
<thead>
<tr>
<th>Parameter / Test method (standard)</th>
<th>Measured value</th>
<th>Requirements (classification) acc. to EN 13162 / Requirements acc. to EN 13500 (ETICS)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Compressive stress (10% deformation) (\sigma_{10}) [kPa] [21]</td>
<td>12.9</td>
<td>(&gt;10kPa) (CS(10)10) / (\geq10kPa)</td>
</tr>
<tr>
<td>Compressibility (c) [mm] [22]</td>
<td>1.79</td>
<td>(\leq2(+1)\ mm) (CP2) / no requirement</td>
</tr>
<tr>
<td>Tensile strength perpendicular to faces (\sigma_t) [kPa] [23]</td>
<td>8.4</td>
<td>(&gt;7.5\ kPa) (TR7.5) / Fixing: - adhesive (\geq80\ kPa) - anchors (\geq7.5\ kPa) - anchored grid (\geq5\ kPa)</td>
</tr>
<tr>
<td>Point load ((\varepsilon=5\ mm)) (F_p) [N] [24]</td>
<td>587</td>
<td>(&gt;550\ N) (PL(5)550) / no requirement</td>
</tr>
<tr>
<td>Shear strength (\sigma_s) [kPa] [25]</td>
<td>28.1</td>
<td>(&gt;28\ kPa) (SS28) / no requirement</td>
</tr>
<tr>
<td>Bending strength (\sigma_b) [kPa] [26]</td>
<td>281.9</td>
<td>(&gt;250\ kPa) (BS250) / no requirement</td>
</tr>
</tbody>
</table>

This article shows that hydrothermal and mechanical performance of developed aerogel-based material meet requirements to be used for external thermal insulation system (ETICS) as well as for internal thermal insulation system. However, important factor for use of such novel insulation as part of insulation system is its durability. Especially critical could be long-term behaviour of aerogel against moisture. The influence of ageing is generally tested in laboratory in scope of material assessment. In this study, ageing tests were performed in wet conditions. The ageing conditions correspond to [27], clause 5.2.4.1.2. In present work, two methods of ageing were used. In method 1, the samples were exposed to heat-moisture actions at 70°C, 95% RH for 7 and 28 days respectively. Afterwards, the aged samples were dried for 7 days at 23°C, 50% RH until constant weight is achieved. In Method 2, the samples were stored over warm water bath. The temperature of water in the container was 60°C. The samples were exposed to vapor only from one side. After 5 days the insulation samples are removed and placed for 28 days in sealed bag at 23°C, followed by 7 days of drying period at

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23°C, 50% RH until constant weight is achieved. The critical material properties, namely tensile strength, thermal conductivity and dimensional stability, were determined for aged insulation. The results are shown in Table 2. [27] does not give any requirement for material behaviour after ageing. According to German approval for mineral wool for use in ETICS, tensile strength after ageing (method 2) must be at least 50% of initial strength (pristine condition). This requirement was fulfilled for each ageing method. The thermal conductivity of the insulation after 1 month of ageing at 70°C, 95% RH (Method 1) is 2 mW/(m·K) higher than for pristine condition. On the other hand, the dimensional stability of samples aged under method 2 was much higher (dimensional stability for length IΔε is eight times higher) compared to pristine condition.

Table 2. Material properties of developed aerogel-based board after ageing.

<table>
<thead>
<tr>
<th>Ageing method / Conditioning</th>
<th>Parameter [unit]</th>
<th>Measured value</th>
<th>Reference value (pristine condition)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Method 1 (7 days heat-moisture actions at 70°C, 95% RH, 7 days drying period at 23°C, 50% RH)</td>
<td>Tensile strength σ1 [kPa]</td>
<td>6.5</td>
<td>8.4</td>
</tr>
<tr>
<td></td>
<td>Thermal conductivity λ [mW/(m·K)]</td>
<td>17.24</td>
<td>17.30</td>
</tr>
<tr>
<td></td>
<td>Dimensional stability IΔε1, IΔε1, IΔε1 [%]</td>
<td>0.05, 0.04, 0.33</td>
<td>0.04, 0.10, 1.09</td>
</tr>
<tr>
<td>Method 1 (1 month heat-moisture actions at 70°C, 95% RH, 7 days drying period at 23°C, 50% RH)</td>
<td>Tensile strength σ1 [kPa]</td>
<td>6.1</td>
<td>8.4</td>
</tr>
<tr>
<td></td>
<td>Thermal conductivity λ [mW/(m·K)]</td>
<td>19.55</td>
<td>17.30</td>
</tr>
<tr>
<td></td>
<td>Dimensional stability IΔε1, IΔε1, IΔε1 [%]</td>
<td>0.09, 0.07, 0.16</td>
<td>0.04, 0.10, 1.09</td>
</tr>
<tr>
<td>Method 2, Series 3 (5 days warm water bath at 60°C, 28 days in sealed bag at 23°C, 7 days drying period at 23°C, 50% RH)</td>
<td>Tensile strength σ1 [kPa]</td>
<td>7.9</td>
<td>8.4</td>
</tr>
<tr>
<td></td>
<td>Thermal conductivity λ [mW/(m·K)]</td>
<td>17.22</td>
<td>17.30</td>
</tr>
<tr>
<td></td>
<td>Dimensional stability IΔε1, IΔε1, IΔε1 [%]</td>
<td>0.32, 0.17, 1.62</td>
<td>0.04, 0.10, 1.09</td>
</tr>
</tbody>
</table>

Material structure of aged samples (Method 1, 1 month at 70°C, 95% RH) was analysed in order to justify material performance after ageing. Surface morphology of the pristine and aged sample was estimated using Analytical Environmental Scanning Electron Microscope (AESEM) Zeiss EVO LS 15. Fig. 5 shows SEM images. The analysis shows breakage of silica network for aged sample.

![Fig. 5. SEM images of developed aerogel-based board (a) pristine material, (b) aged sample.](image)

**Conclusions**

The developed aerogel-based insulation material has a very low thermal conductivity of 17.2 mW/(m·K). It has been shown that the thermal conductivity of a glass fiber insulation material can be reduced by more than half by using aerogel granules.

In addition, the developed composite material is characterized by good hydrothermal properties. The material is both moisture-repellent and permeable to water vapor. Compared to the aerogel-based insulation products available on the market, it can be manufactured more cost-efficient and relatively quickly because of the developed production process.

The material assessment shows that developed aerogel-based insulation board can be used for external thermal insulation system (ETICS) as well as for internal thermal insulation system.
The insulation should be further investigated in detail to some other important characteristics, e.g. VOC emission, fire behavior, dust development, etc. However, for other applications of this promising insulating material, further specific investigations are required.

References

27. ETAG 004:2013. Guideline for European Technical Approval of external thermal insulation composite systems (ETICS) with rendering.