Impact of aeration and deaeration of switchable vacuum insulations on the overall heat conductivity using different core materials and filling gases

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Received: 5 June 2020 / Accepted: 31 August 2020 / Published online: 17 September 2020
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Abstract

Investigating switchable vacuum insulation panels might lead to a new type of insulation, which can be switched on to enable a low heat flow when a good insulation effect is desired and switched off when exchange with the environment is requested, during a cold summer night, for example. For this reason, different core materials for vacuum insulations as typical silica powder were investigated as well as silica agglomerates and silica gel. These materials were checked for the necessary time of aeration and evacuation and the corresponding change of heat conductivity along with the change of gas-pressure. Silica gel in combination with helium as filling gas showed best results corresponding to a high difference of the heat conductivities evacuated and aerated. Beside the solid backbone structure of the silica gel, this is caused by the high heat conductivity and small kinetic atomic diameter of the helium gas. Silica agglomerates decreased the aeration time as well as the deaeration time, but the improvement was neglected because of a lower change of heat conductivity during pressure drop or rise. Nevertheless, a good switchable vacuum insulation can be produced using silica gel and helium, for example.

Keyword Aerated vacuum insulation · deaerated vacuum insulation · switchable vacuum insulation · thermal superinsulation · vacuum insulation · vacuum insulation panels

List of Symbols

a	Parameter [–]
b	Parameter [–]
c	Parameter [–]
d	Parameter [–]
kn	Knudsen number [–]
k	Constant [–]
L	Mean free path of molecules [m]
p	Pressure [Pa]
T	Temperature [°C]
t	Time [s]
x	Pore size [m]
δ	Diameter [m]

Greek

α	Accommodation coefficient [–]
β	Factor [–]
κ	Adiabatic coefficient [–]
λ	Heat transfer coefficient [$\text{Wm}^{-1} \text{K}^{-1}$]
λ	Time-dependent heat conductivity [$\text{Wm}^{-1} \text{K}^{-1} \text{s}^{-1}$]
λ	Pressure-dependent heat conductivity [$\text{Wm}^{-1} \text{K}^{-1} \text{Pa}^{-1}$]
Φ	Porosity

Subscripts

Ae	Aeration
B	Boltzmann
De	Deaeration
end	Ending point
g	Gas
IP	Inflection point
M	Molecule
0	Starting point
10	Logarithm to base ten
∞	Ambient condition

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Introduction

Insulation materials are quite important to reduce the heating and cooling demand of, for example, residential or office buildings and, therefore, to prevent a high-energy demand and thus a high carbon dioxide emission caused by the power generation based on fossil fuels. Besides conventional insulation materials such as Polystyrene, Polyurethane or glass wool, vacuum insulation materials can be a good alternative to improve the energy balance or to reduce heat losses of new or already existing buildings especially when space limitations prevent thick insulations [1].

A disadvantage of those conventional insulation materials is their static behavior when conditions change, such as the outside temperature. In such cases, a switchable vacuum insulation with a large difference in heat conductivity under vacuum conditions and at atmospheric pressure is quite interesting [1, 2]. Some material combinations achieve a difference between aerated with air ($450 \text{ mWm}^{-2} \text{ K}^{-1}$) and evacuated ($35 \text{ mWm}^{-2} \text{ K}^{-1}$) of a factor of around 13. Using hydrogen as filling gas can even lead to a factor of 48 ($1680 \text{ mWm}^{-2} \text{ K}^{-1}$ aerated), whereas improvements of conventional insulations only reach a factor of around 11 and still have the disadvantage of necessary modifications of the existing building structure or expensive new installations [3].

Usually pyrogenic silica is used as filling material for vacuum insulations and is covered with a compound foil consisting of a polymer layer for welding purposes, an alumina layer to prevent any gas leakage or diffusion, and another polymer layer as mechanical protection [4]. This setup has the weakness of not being resilient against mechanical forces and is usually not evacuable after it has been flushed with air, so it has to have a durable impermeability of around 10 kPa during the lifetime of around 50 years [1]. That means that a pressure rise to 10 kPa absolute pressure within the panel is allowed in 50 years. Regularly used fumed silica reaches a heat conductivity of around $3 \text{ mWm}^{-1} \text{ K}^{-1}$ under evacuated conditions and as commercially sold, which means it is filled with opacifier and reinforcing agents and surrounded by a compound foil envelope, of 5 to $8 \text{ mWm}^{-1} \text{ K}^{-1}$ [5, 6]. However, the heat conductivity can change during the lifetime, caused by aging due to high temperatures and thus material degradation or, as is more likely for the building industry, by diffusion effects of gases or water resulting in a reduction of the insulation effect of up to 20% [7]. Such a negative behavior can be prevented when a switchable vacuum insulation is used, which has to be aerated and evacuated during regular operation. A disadvantage when producing a switchable vacuum insulation out of a standard vacuum insulation

panel is the volumetric change when aerated and the loss of mechanical stability. Furthermore, the compact powder mixture is a high flow resistance which leads to a long evacuation time and thus to a very slow change in the heat flow of the wall. This can be improved by installing a gas-drainage system in the form of a non-woven fabric, which is applied flat between the core material and the diffusion barrier, and helps to accelerate the production process of commercial panels, but does not help if the thickness of the panel is increased. However, these problems can be prevented either by using agglomerates or granulates instead of powder or by using gas with a smaller atomic radius for faster aeration and diffusion. At similar pressure conditions, the heat conductivity increases with the grain size from monolithic to powder and finally to granular. The difference in the heat conductivity even increases at higher pressure levels [8]. This effect is caused by different grain sizes causing different nanopores and macropores. The pore size is significantly involved in the total heat flow. On the one hand, the Knudsen Effect, which will be discussed in detail later, is responsible for the thermal conductivity of the gas. On the other hand, it should also be noted that above a certain pore size, when the critical Rayleigh number is exceeded, natural convection will occur in the pores, which should normally be avoided, since the total heat transfer increases due to natural convection. However, this could be an advantage for switchable VIPs [9].

The replacement of air in organic aerogel samples using nitrogen, carbon dioxide, and argon already showed that nitrogen had the highest aeration time, whereas carbon dioxide and argon were slower but similar to each other. Nevertheless, using helium as filling gas showed the highest difference of heat conductivity leading to a conductivity of around $18 \text{ mWm}^{-1} \text{ K}^{-1}$ when aerated [10].

These findings show that an improvement of vacuum insulation towards switchable vacuum insulation panels can be performed by changing the envelope to increase mechanical stability, finding the best grain size for fast evacuation, and the best filling gas for high differences in heat conductivity when aerated and evacuated. Furthermore, these researches can also be of interest using silica as basic material instead, which already shows a low heat conductivity when aerated with air. Thus, the present research shows investigations in the heat conductivity of different grain sizes of precipitated silica under different gas pressure conductions and with various filling gases. Furthermore, the construction design of a switchable vacuum insulation board is shown. This board was then used to test it for evacuation and aeration time measurements for the examined gas–silica combinations and the generating flow resistance.

Materials and methods

Theoretical Background

The main effect, which is responsible for the functionality of vacuum insulations in general, is the reduction of the gas thermal conductivity λ_g inside of the pores of the insulation material. Thereby, λ_g can be significantly reduced by reduction of the gas pressure p due to the Knudsen effect if the mean free path of the gas molecules L is of the same order of magnitude as the pore size x . Equation 1 and Eq. 2 apply [11]. The resulting gas thermal conductivity thus depends not only on the Knudsen number Kn but also on the porosity ϕ and thermal conductivity of the gas under normal conditions λ_∞ :

$$\lambda_g = \frac{\phi * \lambda_\infty}{1 + 2 * \beta * Kn} \quad (1)$$

$$Kn(p, x) = \frac{L}{x} \quad (2)$$

Thereby, the factor β is different for every gas/solid surface combination and can be calculated from the adiabatic coefficient of the gas κ and the thermal accommodation coefficient α (Eq. 3).[11]:

$$\beta = \frac{5\pi}{32} * \frac{9\kappa - 5}{\kappa + 1} * \frac{2 - \alpha}{\alpha} \quad (3)$$

The mean free path of the gas molecule can be calculated out of the Boltzmann constant k_B ($1.38 * 10^{-23} \text{ JK}^{-1}$), the absolute temperature T and the kinetic diameter of the gas molecule δ_M (Eq. 4).[12]:

$$L(p) = \frac{k_B * T}{\sqrt{2} * \pi * \delta_M^2 * p} \quad (4)$$

Reagents

For all tests performed with precipitated silica, the PERKASIL® GT 3000 GRAN obtained from W. R. Grace & Co. was used as basic material. Thereby, it has a maximum pore size distribution at a pore size of about 10 nm [13, 14]. Furthermore, the silica gel Syloid® AL-1 was purchased from W. R. Grace & Co, whose pore size distribution is maximum at a pore size of 3.2 nm [15]. The filling gases used in the experiments were standard laboratory gases neither specially dried nor otherwise processed. Agglomerates were produced out of precipitated silica by mixing it with water in a rotary blender and further drying under vacuum conditions at ambient temperature. The difference in the particle size

Fig. 1 Length fraction of silica powder and mass fraction of silica agglomerates

Fig. 2 Cumulative length fraction of silica powder and mass fraction of silica agglomerates

distribution can be seen in Fig. 1. Because of the quite different order of magnitude of particle sizes, different measuring techniques had to be used to characterize the samples. For the powder materials, the length fraction was determined by microscopy, whereas for the agglomerates, the mass fraction is given by sieve analysis.

Nevertheless, one can recognize that the powder has smaller particle sizes and a smaller distribution. In contrast, the agglomerates are larger and the distribution is more spread (Fig. 2). In the plot, the mass of the agglomerate particles with particle size zero is not zero because it corresponds to the passing of the fine particles that could not be measured.

A 1.15 mm-thick ethylene propylene diene monomer rubber (EPDM) foil, as well as a special adhesive to seal the overlaps, was used as covering material for the switchable vacuum insulation panel (made by Firestone Building Products EMEA). A polymer fleece was purchased at the local construction market and is on the one hand used as a filter to prevent any carry away of the insulation material through evacuation and on the other hand used as a gas-drainage system to ensure an equal pressure distribution over the whole insulation panel. Normal construction wood was used for the frame and was connected with wood screws, both of which were also purchased at a common construction market. The connection to the vacuum pump and the pressure sensor were self-made aluminum bushings, which guaranteed a tight connection to the vacuum pipe system.

Sample analysis

The measurement of the aeration and evacuation was performed using an Ahlborn Almemo® 2490 logger in combination with a Siemens SITRANS P200 (7MF1565-5BB00-1AA1, 0–160 kPa abs.) pressure sensor. The measurement of the heat conductivity was carried out using a self-made guarded hot plate apparatus as mentioned by Sonnicks et al. [13], whereas the same test parameters were used. For this measurement, two separate samples were prepared as a plate or cuboid bulk with dimensions of around 160×160×20 mm and tested simultaneously. Furthermore, each test of the experiment was conducted multiple times. The overall standard deviation includes the errors of each measurement and sensor as well as the deviations of the measurements itself. It is calculated using the Gaussian error propagation law. Uncertainty analysis and error discussion of the used guarded hot plate apparatus can be looked up in the paper published by Sonnicks et al. [13] and is ≤2.07% of the measured value. For the pressure sensor, the number is ≤0.25% of the measured value. It is also shown in Table 1.

Data analysis

The achieved experimental results were further processed with MATLAB® as a mathematical program to generate the necessary balancing functions. The goal of the data analysis was to compare all configurations taking both

thermal conductivity over gas pressure and aeration and deaeration measurements into account. The result is the time-dependent conductivity change during aeration and deaeration. Therefore, the data received such as heat conductivity, pressure, and time were imported into a MATLAB-file and then logarithmized to base 10. Depending on the experiment, different functions were used to fit the data. The respective function was then used to determine the inflection point (IP) of the time-dependent panel pressure at aeration or deaeration processes using the second derivation and the *fzero* function in MATLAB®. This point was chosen, because a full flush of the panel would not be of interest due to the slow deaeration process, and simultaneously, a reproducible point is necessary for comparison reasons.

The point of the beginning of the aeration of the panel could directly be taken from the start of the measurement, whereas the end of the deaeration process was determined using the first derivation and setting the gradient of minus one to zero and using the *fzero* function again. This whole procedure helps to keep the calculations constant and reproducible for all measurements. After the different points were determined, the time steps were then used to determine the pressure values at these points, which are following used to calculate the heat conductivity at these pressure points. The overall schedule for each calculation step of the numerical analysis is shown in Fig. 3.

Having these numbers, the time-dependent (Eqs. (5), (6)) and the pressure-dependent heat conductivity (Eqs. (7), (8)) can be calculated as follows:

$$\lambda'_{Ae} = \frac{\lambda_{IP,Ae} - \lambda_{t=0}}{t_{IP,Ae} - t_0} \quad (5)$$

$$\lambda'_{De} = \frac{\lambda_{IP,De} - \lambda_{t=end}}{t_{IP,De} - t_{end}} \quad (6)$$

$$\lambda'_{Ae} = \frac{\lambda_{IP,Ae} - \lambda_{t=0}}{p_{IP,Ae} - p_0} \quad (7)$$

$$\lambda'_{De} = \frac{\lambda_{IP,De} - \lambda_{t=end}}{p_{IP,De} - p_{end}} \quad (8)$$

These results can show the influence of the pressure drop on the heat conductivity as well as the necessary time to achieve this change. A long evacuation time or a small pressure difference may be compensated by a large drop of the heat conductivity. Preferably, the time-dependent and the pressure-dependent heat conductivities should be as high as possible to enable a big difference between the aerated and the evacuated switchable VIP.

Table 1 Measurement apparatus used in the experiments

Measurement	Apparatus	Uncertainty
Heat conductivity	Guarded hot plate	≤2.07%
Aeration/deaeration	Pressure sensor	≤0.25%

Fig. 3 Schedule of the numerical analysis

Fig. 4 Design and exploded drawing of the switchable vacuum insulation

Experimental setup

As switchable vacuum insulation, a panel was made from a wooden frame covered with EPDM and filled with the corresponding insulation material as shown in Fig. 4. As already mentioned, an overlap of the EPDM layers was used to seal the layers together to ensure a vacuum-tight connection. The whole design has been chosen to be flexible enough to compensate for any volume change during gas flushing of the panel, yet stable enough when evacuated.

The dimensions can be seen in Fig. 5. The connection to the vacuum pump and the aeration unit was on one side while the connection to the pressure sensor was on the other side of the panel as well as in the other corner of the panel. This ensured that the measured pressure was not influenced by the vacuum pump and showed any influences of flow resistances caused by the packed insulation material.

The complete experimental setup is shown in Fig. 6. The vacuum panel is connected to the pressure sensor and

Fig. 5 Design drawing of the switchable vacuum panel with essential dimensions

Fig. 6 Flow chart of the experimental test plant **Table 2** Amount of different materials used for the experiments

Material	Bulk density [kgm ⁻³]	Mass [kg]
Silica powder	200	10.52
Silica agglomerates	356	18.75
Silica gel powder	663	34.88

further on to a computer. The other side is connected to a system of pipes ensuring evacuation of the panel through a vacuum pump and the storage of the gas in a gas bottle compressed by a compressor. The insulation panel can be aerated either from the gas bottle or by opening a valve to the surrounding. To ensure a comparable process a reducing valve is installed behind the gas bottle so the pressure can be reduced to around 101 kPa fitting the same pressure as the surrounding. Verification of the airtightness of the panel as well as of the pipe system with all valves was performed before measurement. The average pressure loss was then calculated to around 400 Pa/h.

The amount of silica and silica gel was calculated to fit a bulk density, which provides the best panel stability in combination with low heat conductivity. As many agglomerates as possible were added to still achieve a flat panel. The bulk density and the absolute masses can be seen in Table 2.

Standard filling gas was the surrounding air, whereby waterless nitrogen was also tested to identify any shifts

Table 3 Heat conductivity (20 °C, 101 kPa) [16] and kinetic atomic or molecular diameter [17] of the used filling gases and of air [14]

Gas	λ [10 ³ Wm ⁻¹ K ⁻¹]	d_{kin} [nm]
Air	26.3	0.36
Nitrogen	25.6	0.364
Helium	153.6	0.26

of the aeration or evacuation process caused by adhering water molecules. Furthermore, helium was chosen as filling gas to see if small molecules accelerate the respective process of filling or draining of the panel. Additionally, helium has a high heat conductivity, which should improve the difference between an aerated and an evacuated vacuum insulation panel. Significant material data of the different gases can be seen in Table 3.

Tests were conducted with the material combinations of silica powder and air or nitrogen, silica agglomerates with air, and silica gel with Air or Helium. As already mentioned was nitrogen gas of main interest to prove if present water molecules attached to the insulation material influence the heat conductivity measurement. Furthermore, Helium was used to see if the high bulk density of silica gel can be improved in case of aeration or deaeration and was not of any interest in combination with agglomerates or powder.

Results and discussion

Heat conductivity

Finding the best combination of filling gas and core material for the switchable vacuum insulation needs the heat conductivity of both materials; the filling and the gas dependent on the gas pressure. All heat conductivities of the investigated combinations are shown in Fig. 7. Silica with air or nitrogen has almost the same heat conductivities for all pressure values. This was to be expected because nitrogen has the most proportion of air. The influence of natural moisture in the air is not in a measurable range. Furthermore, silica gel with air shows a considerably wider spread between heat conductivity at vacuum and atmospheric pressure than precipitated silica powder. This effect is even stronger when the air is replaced with helium, because of the higher gas thermal conductivity of helium compared to air. Considering the thermal conductivities of the gases, which can be found in Table 2, one might expect that the thermal conductivity of silica gel with helium at atmospheric pressure should be even higher. While silica gel/air shows 2.8 times the conductivity of the pure filling gas, silica gel/helium stays around the value of the pure gas. This behavior is caused by several effects. One reason is the smaller molecule diameter d_M of the helium molecule compared to the average of air which leads to a higher mean free path of the gas molecules L and, therefore, to a higher Knudsen number Kn as well as finally to a lower gas thermal conductivity while all other parameters are kept constant. Another reason is the lower thermal accommodation coefficient of helium/silica

compared to air/silica, which leads to a higher β value and thus also a lower heat conductivity [14]. In general, these findings prove a theoretical good possibility as a combination of switchable insulations. Agglomerates still have low heat conductivity and can, therefore, be used as insulation material. They show a reasonable spread of thermal conductivity between minimum pressures and pressure around 1 kilopascal. In contrast to all other samples, the curve seems to drop even further with ongoing pressure reduction. This behavior is due to the macropores created by the agglomeration process. Unfortunately, the pressures required for this can only be achieved at great expense in terms of equipment and is, therefore, not economical for technical applications.

The heat conductivities at the minimum and maximum pressures can be seen in Table 4. Silica gel and helium show the highest ratio of aerated to deaerated and the agglomerates the lowest.

Table 4 Heat conductivity aerated and deaerated and the corresponding ratio

Insulation material/gas	λ_{Ae} [$10^3 \text{ W m}^{-1} \text{ K}^{-1}$]	λ_{De} [$10^3 \text{ W m}^{-1} \text{ K}^{-1}$]	$\lambda_{Ae}/\lambda_{De}$
Silica powder/air	29.4	3.2	9.2
Silica powder/nitrogen	29.4	3.1	9.6
Silica agglomerates/air	41.7	9.1	4.6
Silica gel/air	73.7	3.0	24.4
Silica gel/helium	149.3	3.6	41.1

Fig. 7 Heat conductivity of the experimental gas and insulation combinations

Fig. 8 Aeration of the switchable vacuum panel with different gases and filling materials and starting and inflection points marked (red crosses)

Aeration of the panel

The time-dependent aeration of the panel with different gases and filling materials is shown in Fig. 8. The panel with silica gel and the air is slowest at aeration. Helium improves that behavior because of the mobility of the small molecules. Agglomerates also improve the aeration compared to a packed bulk of silica.

The calculation of the pressure-dependent heat conductivity considering both the heat conductivity and the pressure drop at the starting and the inflection point shows that silica gel in combination with helium produces the best results, followed by silica gel and air (Fig. 9). The agglomerates do not improve the value because the difference of the heat conductivity is smaller than it was possible with silica powder and a higher basic value of the heat conductivity due to a higher starting pressure.

Looking at the time-dependent heat conductivity shows an even better ratio for the combination of silica gel and helium (Fig. 10). This is caused by the fast aeration by the helium molecules and the simultaneously high difference of the heat conductivity. Precipitated silica with nitrogen is slightly better and agglomerates have improved, but both are far from silica gel.

Deaeration of the panel

The results of the experiments when evacuating the insulation panel are shown in Fig. 11. The silica agglomerates have the fastest deaeration time because of the lower flow resistance. The experiments with silica gel show the slowest results because its bulk density is the highest and thus creates a high flow resistance. The evacuation is here only

Fig. 9 Pressure-dependent heat conductivities of different insulation material and filling gas combinations at the aeration process

Fig. 10 Time-dependent heat conductivities of different insulation material and filling gas combinations at the aeration process

caused by the flow resistance because changing to a more powerful vacuum pump has not changed the deaeration time significantly. However, as was already evident during the aeration process, the change from air to helium lowered the time necessary. Furthermore, there is no visible difference when using silica with air or nitrogen showing no sign of the influence of adhered water.

The values calculated from these results are shown in Fig. 12. No big changes to the aeration process are visible. Helium and silica gel are again the best combination.

Compared to the pressure-dependent heat conductivity, the time-dependent heat conductivity shows a difference especially for the agglomerates (Fig. 13). The faster

Fig. 11 Deaeration of the switchable vacuum panel with different gases and filling materials and inflection and ending points marked (red crosses)

Fig. 12 Pressure-dependent heat conductivities of different insulation material and filling gas combinations at the deaeration process

Fig. 13 Time-dependent heat conductivities of different insulation material and filling gas combinations at the deaeration process

evacuation improves the value so that it is higher than silica gel with air. As for all other results, the usage of helium shows the best optimization and gives the highest result. In that case, the combination of agglomerates and helium might be of interest.

Conclusions

Switchable vacuum insulation panels could be an interesting possibility to optimize the temperature profile of a house when the inside temperature is different from the outside one. Depending on the heat sink, either the house or the environment, and the preferences of the residents the heat

flow could be improved or inhibited. Furthermore, when using switchable vacuum insulation panels in electrical devices or technical apparatuses, aerating or evacuating the insulation could help to better control the temperature and improve its functionality and lifetime.

Results of the performed experiments show that fast aeration is only possible using either small gas molecules or big solid particles. Combining the time to aerate the panel with the increase in heat conductivity showed that the fast aeration process of the agglomerates is neglected by the lower difference of the heat conductivity, whereas the time- and pressure-depending heat conductivity of silica gel with helium even increased because of the high difference of the heat conductivities evacuated and aerated. Considering the pressure rise showed that silica gel with air is also not a good combination because of the high bulk density, but can be improved by using helium as filling gas. The results of the thermal conductivity measurements of the tested silica gel in combination with helium are remarkable because a very wide range between the vacuum value and the atmospheric pressure value with a factor of 41.5 could be detected.

The deaeration process depended also mainly on the bulk density that is responsible for the flow resistance. The experiments with silica agglomerates showed the fastest evacuation. However, considering the corresponding change of the heat conductivity as well showed that again silica gel with helium as filling gas was the best combination, followed by silica agglomerates and silica gel with air as filling gas. This means that the fast pressure drop during the evacuation showed that agglomerates could not compensate for the low difference of the heat conductivities.

Overall, the measurements show that no improvement through agglomeration and the corresponding faster air change process can be achieved if the heat conductivity is impaired simultaneously. This means that agglomerates have to be as small as possible or spaces between larger particles have to be filled with smaller ones. If then combined with a filling gas such as helium, which offers a large spread of the thermal conductivity between the two switching states of the panel, good switchable vacuum insulation can be produced mainly for technical applications.

Acknowledgements This work was funded by the German Federal Ministry for Economic Affairs and Energy (AiF Project GmbH, Funding Code: ZF 4013937CL7). All authors declare that they have no conflict of interest whatsoever.

Funding Open Access funding enabled and organized by Projekt DEAL.

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