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Thermal conductivity of vacuum insulation materials for thermal energy stores in solar thermal systems

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Abstract

The effective thermal conductivity of fine-grained and coarse-grained expanded perlites, fumed silica and a mixture of coarse-grained expanded perlite and fumed silica at different vacuum pressures is experimentally investigated in this paper. For the investigations two different test rigs – a guarded cylinder apparatus and a guarded hot plate apparatus – were used. The lowest effective thermal conductivities were performed with fumed silica at ambient pressure, with the mixture at pressures between 1 and 10 mbar and with fine-grained expanded perlite at 0.1 mbar and lower. Mixtures of fumed silica and expanded perlites appear to be a great chance to achieve low effective thermal conductivities and lower prices compared to pure fumed silica at vacuum pressures that can be easily achieved for large-volume thermal energy stores.

1. Introduction

To achieve high solar fractions in solar thermal applications, high efficient hot water stores with high heat capacities and thereby large volumes can be necessary. However installing such stores into dwellings means a significant reduction of living space. Furthermore the heat losses of the store in summer can lead to an unrequested raise of temperature in the dwelling. An outdoor installation of thermal energy stores (TES) can solve both

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problems. Lower ambient mean temperatures outdoor compared to indoor however increase the significance of a high efficient thermal insulation. Even indoor utilization of high efficient insulated TES can reduce both problems by decreasing the heat losses in summer (and thus the unrequested raise of temperature in the dwelling) and by decreasing the necessary heat capacity and volume of the TES [1]. Vacuum insulation can reach the lowest effective thermal conductivity. Thus investigations are carried out to develop and improve this technology for hot water stores [2]. This contribution shows measuring results of the effective thermal conductivity of suitable insulation materials, such as newly developed mixtures, for vacuum insulation. Furthermore the built up test rigs will be described.

Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>GC</td>
<td>Guarding cylinder</td>
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<tr>
<td>GCA</td>
<td>Guarded cylinder apparatus</td>
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<tr>
<td>GHD</td>
<td>Guard heater deviation</td>
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<tr>
<td>GHPA</td>
<td>Guarded hot plate apparatus</td>
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<tr>
<td>MH</td>
<td>Measuring heater</td>
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<tr>
<td>TES</td>
<td>Thermal energy store</td>
</tr>
<tr>
<td>ΔT</td>
<td>Temperature difference [K]</td>
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<tr>
<td>λ\text{cpl}</td>
<td>Thermal conductivity due to the coupling effect [W/(m·K)]</td>
</tr>
<tr>
<td>λ\text{eff}</td>
<td>Effective thermal conductivity [W/(m·K)]</td>
</tr>
<tr>
<td>λ\text{gas}</td>
<td>Thermal conductivity of gas [W/(m·K)]</td>
</tr>
<tr>
<td>λ\text{r}</td>
<td>Thermal conductivity due to heat transfer via infrared radiation [W/(m·K)]</td>
</tr>
<tr>
<td>λ\text{s}</td>
<td>Thermal conductivity of the solid matrix [W/(m·K)]</td>
</tr>
<tr>
<td>l\text{HC}</td>
<td>Length of heating cylinder [m]</td>
</tr>
<tr>
<td>Q</td>
<td>Heat flux [W]</td>
</tr>
<tr>
<td>Q\text{S}</td>
<td>Heat flux through the sample container (including the sample) [W]</td>
</tr>
<tr>
<td>Q\text{Ref}</td>
<td>Heat flux through the reference material [W]</td>
</tr>
<tr>
<td>S</td>
<td>Shape factor [m]</td>
</tr>
<tr>
<td>S\text{GCA}</td>
<td>Shape factor of the guarded cylinder apparatus [m]</td>
</tr>
<tr>
<td>S\text{HC}</td>
<td>Outer diameter of the heating cylinder [m]</td>
</tr>
<tr>
<td>S\text{VC}</td>
<td>Inner diameter of the vacuum chamber [m]</td>
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2. Basic principles

In insulation materials usually three different heat transfer mechanisms take place: thermal conduction of solids and fluids, heat flux via infrared radiation and convection. Another effect that increases the effective thermal conductivity of primarily fiber materials and pourable particles (fillings) is the coupling effect. This describes that (residual) gas molecules tend to accumulate near the contact spots of fibers or particles and increase the local thermal conductivity around theses spots. At atmospheric pressure this coupling effect can be responsible for up to 50 % of the effective thermal conductivity of fine grained fillings. Convection can be neglected for the insulation materials considered in this paper because of their small effective pore sizes. The other heat transfer mechanisms mentioned above, can be described as thermal conductivities which can be summed up to the effective thermal conductivity $\lambda_{\text{eff}}$ [W/(m·K)] (see equation (1)). This superposition principle has been established successfully in many scientific works (e. g. [3-8]).

$$\lambda_{\text{eff}} = \lambda_{\text{r}} + \lambda_{\text{s}} + \lambda_{\text{gas}} + \lambda_{\text{cpl}}$$

- $\lambda_{\text{r}}$: thermal conductivity due to heat transfer via infrared radiation [W/(m·K)]
- $\lambda_{\text{s}}$: thermal conductivity of the solid matrix [W/(m·K)]
- $\lambda_{\text{gas}}$: thermal conductivity of gas [W/(m·K)]
\( \lambda_{\text{cpl}} \) thermal conductivity due to the coupling effect [W/(m·K)]

\( \lambda_r \) increases with the temperature and decreases with the insulation materials’ increasing capability to extinct (absorb or reflect) infrared radiation and with increasing bulk density. \( \lambda_s \) increases with the bulk density, the external pressure and the thermal conductivity of the solid the solid matrix is made of and it decreases with increasing porosity. \( \lambda_{\text{gas}} \) increases with vacuum pressure, the effective pore size of the porous material and with the temperature. The smaller the pores of the insulation material are, the higher the vacuum pressure can be to eliminate \( \lambda_{\text{gas}} \) (see Fig. 1).

Fig. 1. Thermal conductivity of air in dependence of the vacuum pressure, calculated with a mathematical model of Schwab [4] for different gaps or effective pore sizes (0.001 mm ... 1 mm).

Increasing the bulk density of a free flowing material leads to smaller pores and thus can lead to a decreasing thermal conductivity of gas. \( \lambda_{\text{cpl}} \) increases with the vacuum pressure, the temperature and the numbers of contact spots between the fibers of fiber mats or between particles of fillings.

3. Methodology

3.1. Measurements

For measuring the effective thermal conductivity there are two different test rigs. One consists of a guarded hot plate apparatus (GHPA) and a sample container for insulation materials wherein the vacuum pressure can be adjusted (see Fig. 2a). The stakes placed into this vacuum sample container are necessary to withstand the load resulted by the vacuum pressure. Thus their influence on the measurement results needs to be calculated. Fig. 2b shows the measurement principle of the GHPA.

The other test rig is a guarded cylinder apparatus (GCA) consisting of a cylindrical vacuum chamber that contains a concentrically placed heating cylinder with guarding cylinders (see Fig. 3). The vacuum chamber can be heated separately and it can be filled with pourable insulation materials.

Both test rigs work according to the same steady state principle: a predefined temperature difference (\( \Delta T \) [K]) is set to the sample. The inner heating element (heating plate or heating cylinder) is set to the higher temperature. As the inner heating element runs electrically the heat flux \( \dot{Q} \) [W] can be measured using a power meter. Taking the geometrical dimensions into account (surface and thickness of the measuring area of the sample, included in a shape factor \( S \) [m]), the effective thermal conductivity \( \lambda_{\text{eff}} \) [W/(m·K)] can be calculated according to equation (2).

\[
\dot{Q} = S \cdot \lambda_{\text{eff}} \cdot \Delta T
\]  

(2)
Fig. 2. (a) vacuum sample container (opened); (b) measuring principle of the GHPA. A: measuring hot plate, B: guarding heat ring, C: cooling plates, D: temperature measurement, E: sample container (including sample), F: reference material with known effective thermal conductivity, G: mats to ensure proper contact between the plates and the samples, \( Q_S \): heat flux through the sample container (including the sample), \( Q_{Ref} \): heat flux through the reference material.

The shape factor of the GHPA is the quotient of the measuring hot plates’ area (multiplied by two for symmetrical measurements) and the thickness of the sample. The first calculation of the GCAs’ shape factor \( S_{GCA} \) [m] is described in equation (3).

\[
S_{GCA} = \frac{2 \cdot l_{HC}}{\ln(s_{VC}/s_{HC})}
\]

\( l_{HC} \) Length of heating cylinder [m]
\( s_{HC} \) Outer diameter of the heating cylinder [m]
\( s_{VC} \) Inner diameter of the vacuum chamber [m]

However equation (3) is only valid for ideal elimination of any edge effects. Because the edge effects cannot be eliminated totally by the guarding cylinders and because of some necessary cables inside the measuring area the shape factor is determined via calibration measurements. Therefore measurements were carried out in the GCA at atmospheric pressure with a material previously measured symmetrically in the GHPA in open sample containers at the same conditions (same temperature, same bulk density, material stored before at the same climatic conditions). Thus the shape factor could be adjusted to meet the same results measured in the GHPA. Results of measurements with other materials in the GCA showed deviations of 10 to 11% to the results generated in the GHPA with the same materials in open sample containers and under the same conditions. Thus the error of the shape factor on the measured effective thermal conductivity is estimated with ±11%.

To determine the influence of the vacuum sample container for the GHPA on the measurement results several measurements, calculations and simulations were carried out. The effective thermal conductivity of several insulation materials were measured in the GHPA with and without the vacuum sample container at atmospheric pressure. Furthermore there were measurements with and without the removable stakes inside the vacuum sample container done. Whereas the influence of the stakes was small and could be determined quite simple with the measurements and calculations, there was still another phenomenon highly affecting the measurement results. Even very small temperature deviations (0.1 to 0.5 K) between the measuring hot plate and the guarding heat ring (guard heater deviation; GHD) can lead to large errors due to the high thermal conductivity of the vacuum sample containers’ upper shell that is applied to the hot plate and guarding heat ring. Thus additional temperature measurements on the hot plate and the guard ring with calibrated Pt100 sensors and thermal simulations were carried out to determine the influence of these GHD. However it was not possible by now to determine the error sufficient, because the mean temperature of the temperatures measured on several spots on the hot plate and the
guarding heat ring cannot be assumed to represent the whole boundary area between. Measurements without the vacuum sample container in the GHPA however are very reliable, proven by many measurements with reference materials. Thus results of the materials at atmospheric pressure can be used to calibrate the GCA. The maximum error of the GHPA can be estimated with ±5 %.

In opposition to the GHPA in the GCA the GHD can be adjusted manually. Even though the temperature field between the heating cylinder and the guarding cylinder is also not known exactly, with the results of the GHPA at atmospheric pressure and with finding a linear correlation between GHD and the measurement results the error is estimated comparatively small. With an assumption of max. ±0.5 K deviation between measured and actual temperature difference the maximum error is estimated with ±1.4 mW/(m·K).

Fig. 3. (a) functional sketch of the GCA ($l_{HC}$: length of heating cylinder, $s_{VC}$: inner diameter of the vacuum chamber, $s_{HC}$: outer diameter of the heating cylinder; (b) heating cylinder and guarding cylinders (separated by black thermal resistances) with temperature sensors and mounting; (c) vacuum chamber with heating tape; (d) installed GCA (covered in thermal insulation) with EMSR technology and vacuum equipment.
3.2. Insulation materials

The application for the vacuum thermal insulation considered in this paper is a double-walled hot water store. The insulation material is filled in the gap between the walls, which is later evacuated. As the outer wall is rigid and sustains the load resulted by the vacuum pressure, the insulation material is not exposed to an external load, unlike it is in vacuum insulation panels. There are mainly two reasons why the gap is filled with insulation materials. The main reason is – as seen in Fig. 1 – that the thermal conductivity of air and the coupling effect can be eliminated only with very low vacuum pressures for big gaps or pores. However it is very extensive to create and maintain that low vacuum pressure. With filling porous insulation materials with small and open pores into the gap the vacuum pressure to eliminate the thermal conductivity of air and the coupling effect, which have the biggest share on the effective thermal conductivity of nearly every insulation material at ambient pressure, can be set to higher values. Furthermore the heat transfer via radiation can be decreased by the insulation material. The heat transfer via the solid matrix that is applied with the insulation material however has a comparatively small share on the effective thermal conductivity of most insulation materials.

The main requirements to insulation materials for the named purpose are:

- Pourable (as they need to be applied in the difficult to access gap of the described double wall)
- Open-pored (as the pores need to be evacuated)
- No outgassing substances (to maintain the vacuum)
- Temperature resistant
- Small sized pores
- High extinction capability of infrared radiation
- Low cost

In this paper the insulation materials expanded perlites (coarse-grained and fine-grained) and fumed silica are investigated. Perlites are amorphic volcanic glasses that contain crystallization water. To create expanded perlite fillings the perlites are powdered and heated to temperatures between 850 °C and 1000 °C. Thus the perlites get soft and the bounded water evaporates, creating small pores and thereby expands the perlite particles. The perlites investigated in this paper have maximum grain sizes of 2 mm (coarse-grained, see Fig. 4a) and 200 μm (fine-grained, see Fig. 4b).

Fumed silica is made from flame pyrolysis and consists of pure SiO₂. The primary particles of the fumed silica investigated in this paper have grain sizes of 10 to 15 nm. These primary particles are arranged to fibrous aggregates with equivalent diameters of 100 to 150 nm. These aggregates build agglomerates with equivalent diameters of around 10 μm. Under slight external pressure these agglomerates can build big clusters with a high porosity (see Fig. 4c). Fumed silica is the mostly used core material for vacuum insulation panels. Therefore it is mixed with fibers and infrared opacifiers – due to its poor capability of infrared extinction compared to perlites – and compressed to boards.

![Fig. 4. (a) coarse-grained expanded perlite; (b) fine-grained expanded perlite; (c) fumed silica [9].](image-url)
With fumed silica much smaller effective pore sizes can be reached than with expanded perlites. However the price of fumed silica is with 4.5 to 8 €/kg [10] much higher than for expanded perlite with ca. 1.2 €/kg [11]. To decrease the pore sizes between the grains of coarse grained perlite it appears to be promising to add a smaller amount of fumed silica. Thus a mixture of 70 mass-% coarse grained perlite and 30 mass-% fumed silica is investigated in this paper.

4. Results and discussion

The results of the effective thermal conductivity as a function of vacuum pressure measured in the GCA at a mean temperature of 48 °C and a temperature difference of 10 K between heating cylinder and vacuum chamber are presented in Fig. 5.

![Fig. 5. Effective thermal conductivity as a function of vacuum pressure measured in the GCA at a mean temperature of 48 °C and a temperature difference of 10 K between heating cylinder and vacuum chamber. The bulk density of each insulation material can be found in the caption in parentheses.](image_url)

The effective thermal conductivity of fine-grained expanded perlite at 970 mbar (ambient pressure at the investigation site) is used as reference and matches the value measured in the GHPA. Thus the max. measurement error on this value correlates to the max. error of the GHPA with ±5 %. As mentioned in chapter 3.1 the error on the other measuring points can be estimated with ±11 % on the effective thermal conductivity for the shape factor calibration and with max. ±1.4 mW/(m·K) due to uncertainties of the determination of the GHD.

The fine-grained expanded perlite has higher effective thermal conductivity at atmospheric pressure (61.6 mW/(m·K)) than the coarse-grained expanded perlite (46.4 mW/(m·K)). This can be explained mainly by a higher coupling effect due to more contact points between the finer grains. The higher bulk density also leads to a higher thermal conductivity of the solid matrix, but as the effective thermal conductivity at 0.02 mbar is lower for the fine-grained expanded perlite (8.3 mW/(m·K) vs. 11.5 mW/(m·K)) the effect of decreasing the radiative heat flux with higher bulk densities seems to overcompensate the increase of solid matrix heat flux. As the effective thermal conductivity of the coarse-grained type is not constant between 0.1 and 0.02 mbar influences of residual gas cannot be excluded, as well as the described measurement uncertainties. Furthermore the thermal conductivity of air in the
fine-grained expanded perlite can be decreased and eliminated with higher vacuum pressures due to the smaller pores – especially between the grains – compared to the coarse-grained type. In the literature there are with theoretically 7 mW/(m·K) lower values found for the same type of coarse-grained expanded perlite at 0.01 mbar and 50 °C with a bulk density of 60 kg/m³ [12].

Fumed silica has with 34.6 mW/(m·K) the lowest effective thermal conductivity of the investigated materials at ambient pressure. That can be explained by the very low thermal conductivity of the solid matrix – due to the low bulk density and high porosity – and with the very small effective pore size, decreasing the thermal conductivity of air even at atmospheric pressure. Because of the small pore size fumed silica reaches its’ lowest effective thermal conductivity at the highest vacuum pressure compared to the other materials. However the effective thermal conductivity at low vacuum pressures is with 16.4 mW/(m·K) noticeable higher compared to the expanded perlites. This can be explained with the lower infrared extinction of fumed silica with a Rosseland mean extinction coefficient of 23 m²/kg at 25 °C [3] compared to expanded perlites with ca. 43 m²/kg at 26 °C [12] and with the lower bulk density also increasing the heat flux via infrared radiation. Because of the poor infrared extinction of fumed silica it is commonly mixed with an infrared opacifier that can increase the Rosseland mean extinction coefficient to 90 m²/kg at 25 °C [3]. To check the plausibility of the measured value of fumed silica the thermal conductivity due to heat transfer via infrared radiation can be calculated according to the gray Rosseland approximation, e. g. described in [13]. With a mean temperature of 48 °C, a bulk density of 43 kg/m³ and the assumption that the Rosseland mean extinction coefficient of 23 m²/kg is also valid at 48 °C mean temperature, a value of 10.1 mW/(m·K) can be calculated. That would imply a thermal conductivity of the solid matrix of 6.3 mW/(m·K), which is very high compared to the literature, where values of lower than 1 mW/(m·K) are described for fumed silica at a bulk density of 50 kg/m³ and mixed with opacifiers that have higher solid thermal conductivities than fumed silica [3]. As the measured values for the coarse-grained expanded perlite are also higher than described in the literature (see above) further measurement uncertainties that are not considered yet cannot be excluded.

The mixture of coarse-grained expanded perlite and fumed silica seems to be a promising chance to reduce both the effective thermal conductivity compared to perlites at vacuum pressures of around 10 mbar and the costs compared to pure fumed silica. The mixtures’ effective thermal conductivity is with 38.2 mW/(m·K) higher compared to fumed silica at ambient pressure – due to the larger effective pore sizes leading to a higher thermal conductivity of air and due to the higher bulk density leading to a higher thermal conductivity of the solid matrix. However the expanded perlite decreases the heat flux of infrared radiation and thus the effective thermal conductivity is lower with vacuum pressures of 10 mbar and beneath compared to fumed silica.

Achieving and maintaining vacuum pressures of 1 mbar and lower at mean temperatures in the insulation of hot water stores with around 50 °C turned out to be very challenging for large volumes of fine porous materials. Thus finding a low-cost material that offers low effective thermal conductivities at easy to achieve vacuum pressures is a very important task to promote the technology of vacuum thermal insulated systems like thermal energy stores. Further experiments with smaller shares of fumed silica or with the lower priced precipitated silica will be carried out during the further procedure.

Fig. 6a shows the effective thermal conductivity as a function of the samples’ mean temperature, measured in the GHPA at atmospheric pressure for fine-grained and coarse-grained expanded perlites and for fumed silica. While the inclination of effective thermal conductivity with the temperature is constant and nearly equal for the perlites, the fumed silica’s inclination is higher and increases with higher temperatures. That shows the lower infrared extinction compared to the perlites described above. Because of the higher bulk density of fine-grained perlite compared to the coarse-grained type a higher inclination of the coarse-grained type – due to higher heat transfer caused by infrared radiation – may be expected. However the difference of those thermal conductivities (calculated with the gray Rosseland approximation described in [13]) is with max. 2.6 mW/(m·K) (for 86 °C) small compared to the effective thermal conductivity and is thus within the measurement error of the GHPA.

An utility model of a vacuum insulated hot water store with 12 m³ water content, 70 mass-% coarse-grained expanded perlite and 30 mass-% fumed silica as insulation material and a transparent thermal insulation (see Fig. 6b) was built up by the company Sirch Tankbau-Tankservice-Speicherbau GmbH and tested within the project “StoEx” (see chapter “Acknowledgements”). A first measurement was done to investigate the mean temperature drop of the storage medium in periods of no charging and discharging. During the 17 days of measurement the
vacuum pressure in the vacuum thermal insulation was approximately 1.1 mbar, the mean temperature of the storage medium was 57 °C and the mean temperature difference between the storage medium and the ambient was 38 K. Only a small overall mean temperature drop of the storage medium of approximately 0.25 K per day occurred. Considering only the upper half of the prototype an even smaller mean temperature drop of approximately 0.1 K per day occurred due to the fact that all thermal bridges of the TES (support construction, pipework and manway opening) are at the bottom.

Fig. 6. (a) Effective thermal conductivity as a function of the mean temperature, measured in the GHPA at atmospheric pressure and a temperature difference of 10 K between hot plate and cooling plates. The bulk density of each insulation material can be found in the caption in parentheses; (b) Utility model of a vacuum insulated thermal energy store for outdoor installation with transparent thermal insulation.

5. Conclusions and outlook

The effective thermal conductivities of four different vacuum insulation materials at 5 to 6 different vacuum pressures were investigated in this paper. Measurements were carried out in two different test rigs. The guarded cylinder apparatus could produce more reliable data for pressures lower than ambient. The guarded hot plate apparatus was used to produce reliable data at ambient pressure and mean temperatures between 10 and 86 °C. Fumed silica has the lowest effective thermal conductivity at ambient pressure and small-grained expanded perlite at low vacuum pressures of 0.1 mbar and beneath. The most interesting vacuum pressures for large thermal energy stores in solar thermal applications are 1 mbar and higher as experiences with an utility model of a TES shows, that lower vacuum pressures are very hard to achieve at high temperatures. At vacuum pressures between 1 and 10 mbar a mixture of 70 mass-% of coarse-grained expanded perlite and 30 mass-% of fumed silica shows the best results. This mixture was also applied to the utility model TES whereby excellent results of temperature drop measurements could be achieved. As fumed silica is considerably higher priced than expanded perlites, mixtures with smaller shares of fumed silica or with the lower priced precipitated silica will be investigated during the further procedure. Furthermore, as the infrared extinction of silica is rather poor, silica will be mixed with infrared opacifiers. Some measurement values – especially at low vacuum pressures – are higher than expected by literature values or theoretical considerations. Thus both test rigs will be optimized and further effort will be done to investigate the measurement errors.
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