

Influence of Water on the Total Heat Transfer in ‘Evacuated’ Insulations

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1 Introduction

Different kinds of filler materials, such as glass fibre boards, pressed powder boards or special open porous and thus evacuable foams are used as core material for Vacuum Insulation Panels (VIPs) [Heinemann 1999]. Least requirements to the quality of the vacuum, that has to be achieved and maintained, is found for nanostructured materials. For example in boards made of pressed fumed silica even at an internal gas pressure of some hundreds Pascal the contribution of the gas to the total heat transfer is nearly completely eliminated [Caps 2001]. For these extremely fine structured core materials also the requirements to the tightness of the envelope are relatively moderate and thus special high barrier metalized laminates consisting mainly of polymers and extremely thin Al-layers can be used for the envelope. As for all plastic films also for these high grade films the transmission rate for water vapour is several orders of magnitude larger than those for oxygen or nitrogen [Schwab 2005]. Thus the impact of water vapour on the thermal conductivity was investigated and found to be surprisingly large. We pursued the question whether not only water vapour within the pore space, but also H_2O in the adsorbed phase contributes to the total heat transfer. To quantify these effects several series of thermal conductivity measurements were performed in an evacuable guarded hot plate [Heinemann 1995]. Aim of the investigations was to formulate an analytical equation that describes the influence of water vapour and adsorbed water on the total heat transfer.

The fumed silica specimens were first investigated in the dry state. Then water vapour was let into the vacuum system in different quantities (valves closed, no pumping), yielding a water content in the VIP-kernel of up to 4% by weight. Most of the water is adsorbed in the specimen, smaller amounts are found in the gas phase. The parameter varied was the temperature and thus accordingly the partial pressure of water vapour.

2 Thermal transport in porous insulations

2.1 Heat transfer in dry insulation materials

In highly porous insulation materials such as foams, fibre-boards or pressed powder boards, total heat transfer occurs via conduction along the solid structure, conduction of the gas within the hollow spaces and infrared radiative transfer. Convection, a very efficient heat transfer mechanism, usually is suppressed in insulation materials used at ambient conditions with moderate temperature gradients. In a simple approach the total thermal conductivity λ can be described as the sum of the gaseous conductivity λ_{gas} , the solid conductivity λ_{sc} and the radiative conductivity λ_{rad} . As this simple approach implies independent temperature characteristics for each heat transfer path, local coupling of the

different mechanisms results in a more or less increased total conductivity. These effects may be accounted for in an additional coupling term λ_{coupl} [Fricke 1993]:

$$\lambda = \lambda_{\text{gas}} + \lambda_{\text{sc}} + \lambda_{\text{rad}} + \lambda_{\text{coupl}} \quad . \quad \text{Equation (1)}$$

Significant coupling effects have been observed in semi-transparent media like monolithic aerogels or low density foams in the combination with low emissivity boundaries or integrated radiation shields [Heinemann 1996], [Caps 1997], as well as in materials with dot like contacts between fibres or grains [Kaganer 1969]. E.g. in a bed of glass spheres, comparing the total thermal conductivity of the evacuated and the non-evacuated specimen, the contribution from the gas is seven times as large as expected just from the thermal conductivity of still air [Heinemann 1995]. In both cases for a single heat transfer mechanism there are localised steep temperature gradients which are related to high thermal resistances. These thermal resistances may be shorted out by other mechanisms.

In optically thick foams with a non-broken structure coupling effects may be neglected. In the evacuated case in optically thick media total conductivity λ_{evac} is:

$$\lambda_{\text{evac}} = \lambda_{\text{sc}} + \lambda_{\text{rad}} \quad . \quad \text{Equation (2)}$$

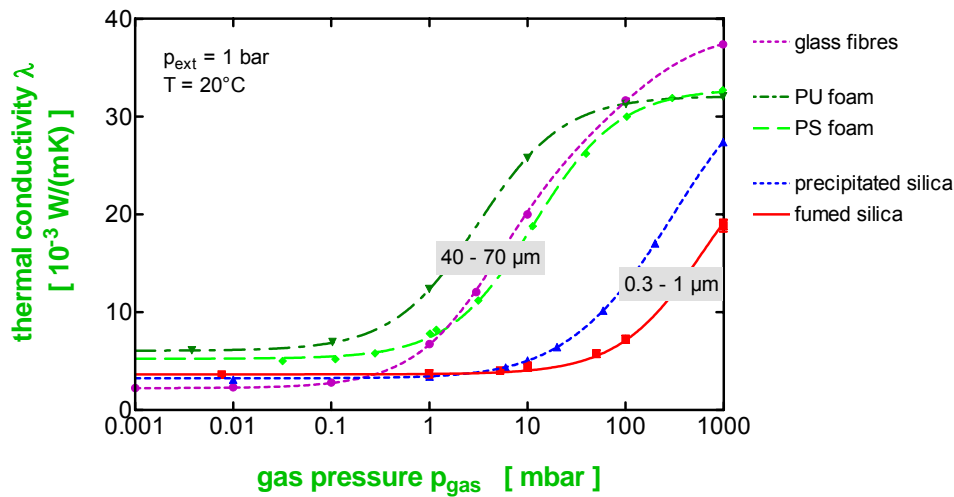


Figure 1: Thermal conductivity of different filler materials more or less optimized for the application in vacuum insulation panels. External pressure is 1 bar, the temperature 20°C.

The variation of the gaseous conductivity with gas pressure p depends on the size of the (largest) pores:

$$\lambda_{\text{gas}} = \lambda_{\text{gas},0} / (1 + 2\beta \cdot Kn) = \lambda_{\text{gas},0} / (1 + p_{1/2} / p), \quad \text{Equation (3)}$$

with $Kn = l/\Phi$ the Knudsen number, with l being the mean free path of the gas molecules and Φ the pore width. $\lambda_{\text{gas},0}$ is the thermal conductivity of the free still gas, β a constant which beside other factors is determined by the accommodation coefficient α [Kaganer 1969]. α describes the efficiency of energy transfer when gas molecules hit the solid structure. It depends on temperature and on the collision partners. For air at 300 K $\lambda_{\text{gas},0} = 0.026 \text{ W/(m·K)}$ and $\beta \approx 1.5$ holds. $p_{1/2}$ is the gas pressure at which the gaseous thermal conductivity is equal to $\lambda_{\text{gas},0}/2$. For a given temperature and gas it is a characteristic property of the insulation material. It may be derived from thermal conductivity measurements as depicted in fig. 1.

Looking at the temperature dependence of the total thermal conductivity, temperature dependencies of all contributions have to be considered: The radiative contribution varies with the third power of absolute temperature, if the extinction coefficient is temperature independent. For amorphous materials at ambient conditions the temperature dependency of the solid conduction can be considered to be the same as for the bulk material. For the gaseous contribution for ideal gases $\lambda_{gas,0} \propto \sqrt{T}$ and $l \propto T$ holds. More precise considerations also take into account spectral variation of the infrared properties and variations in the accommodation coefficient.

Considering the influence of other gases on the total heat transfer, modified parameters for the conductivity of the still free gas $\lambda_{gas,0}$, another mean free path l and another constant β , possibly also with another accommodation coefficient α , have to be taken into account.

2.2 Heat transfer in moist porous materials

The modelling of the coupled heat and moisture transport in porous materials is a major research topic especially in building physics. In contrast to heat conduction, there is no universal theory for the driving forces and transport parameters in the case of moisture transport [Rudtsch 2000]. Besides influences on the parameters describing the heat transfer of the dry material, i.e. likely an reduced infrared radiative transfer, an increased 'solid' conduction by the (non-moving) liquid film, especially when moisture is adsorbed at thermal sensitive dot like contacts and an modified gaseous contribution (see above), the diffusion of vapour in the gaseous phase and of water along the surface have to be considered. Also in the hygro-thermal steady-state which may be achieved in encapsulated insulating materials (as in VIPs), where there is no net moisture transport through the panel, significant or even dominant additional contributions to the total heat transfer may arise from permanent de- and adsorption of water in the hot respectively cold region of the specimen and counterbalanced flow in the liquid and the gaseous phase. Due to large amounts of latent heat from phase changes this so-called heat pipe effect is a very efficient heat transfer mechanism, and thus should be avoided in thermal insulations.

2.3 Measurement method

Thermal conductivity experiments on moist materials are complicated by the additional transport processes of water de- and adsorption and vapour, surface and capillary diffusion. Suitable for the measurement of moisture effects on heat transfer is a long-term observation with a guarded-hot-plate or a heat flow-meter apparatus (see e.g. [Langlais 1983]).

Such thermal conductivity measurements in principle require a temperature gradient, which causes a moisture redistribution in the material. When thermal and moisture equilibrium are reached, moisture transfer by vapour diffusion is counterbalanced by surface back-transfer.

Depending on the water content and on the adsorption characteristic of the material also non-linear moisture profiles have to be taken into account. Thus one has to consider that the measured thermal conductivity is an apparent one.

3 Investigations on 'evacuated' fumed silica

3.1 Investigated material

Boards made by compression of fumed silica powder, mixed with organic fibres for reinforcement and some infrared opacifier proved to be a good filler material for vacuum insulation panels [Caps 2001].

Subject of the investigations described below were such boards, produced in the 3rd quarter of 2002 by the company Wacker Chemie GmbH / Kempten, Germany, labelled by WDS-NT.

The density of the material, loaded with an external pressure of 1 bar was 170 kg/m³.

A first set of measurements was performed in November 2003, a second set in spring 2005. In the meantime the samples were stored in the lab under almost constant conditions.

Measurements of the thermal conductivity on the same type of material, performed on the preheated, dry material were reported e.g. in [Caps 2001]. First investigations on the influence of water on the thermal conductivity on specially prepared vacuum insulation panels are described in [Schwab 2005].

3.2 Sorption Characteristic

The sorption isotherm was measured by the company Quantachrom/Odelzhausen, Germany on the material Wacker WDS-VIP based on fumed silica of the type T30 with a specific surface of about 300 m²/g. It is assumed that there is no significant difference in the sorption behaviour for the materials labelled WDS-NT and WDS-VIP. Initially the material was dried under vacuum for 3 hours at 150°C. The sorption isotherm then was determined for an adsorption and subsequent by a desorption cycle at a temperature of 23°C.

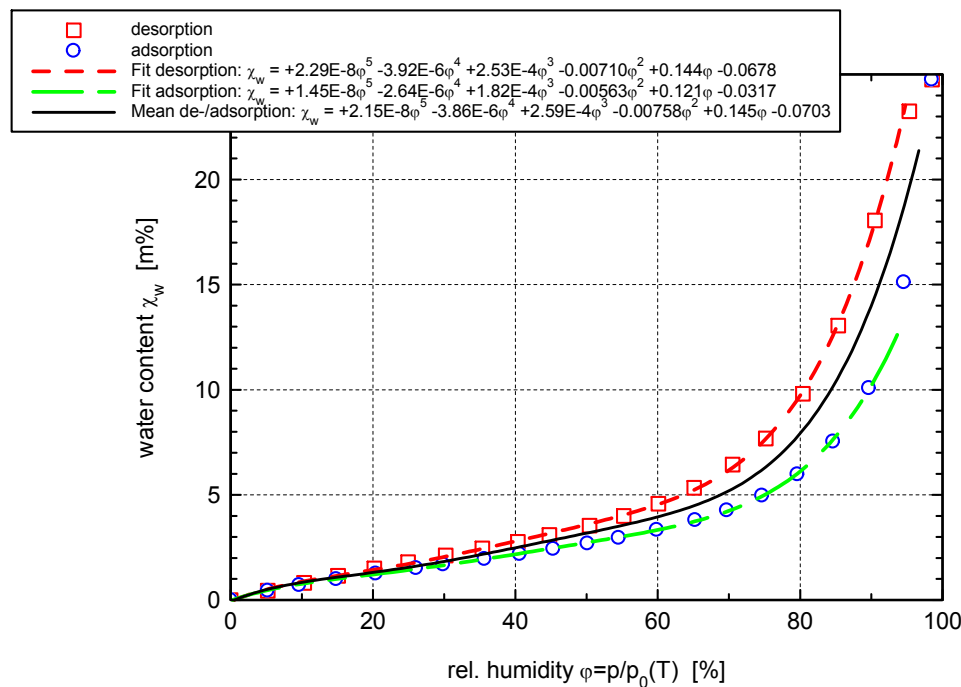


Figure 2: Sorption isotherm for Wacker WDS-VIP derived at 23°C. Measured data for the adsorption and the desorption cycle are depicted together with fit equations used for numerical calculations (see text). Measurements by Quantachrom

As the material investigated is meso/macroporous no micro-pore condensation is observed. Water content X_w and relative humidity ϕ correlate in a linear fashion,

$$X_w = 0.08 \text{ m\%} \cdot \phi / \%, \quad \text{Equation (3)}$$

up to $\phi = 60\%$. For higher humidity polynomial approximations may be used. From the inverse function $\phi(X_w)$ of the sorption isotherm $X_w(\phi)$ one can determine the water vapour pressure p_{wv} for a

given water content in the material. As the saturated vapour pressure increases exponentially with temperature, one gets:

$$p_{wv} = \varphi(X_w) \cdot p_{wv,saturated}(T) \quad . \quad \text{Equation (4)}$$

The adsorption isotherm is assumed to be almost independent on temperature.

3.3 Modelling of the moisture distribution in samples with temperature gradient

In a zero order approximation it was assumed that in thermal and hygric steady-state there is no vapour diffusion or surface diffusion. Thus locally From this results a constant water vapour partial pressure all over the material. According equation (4) the relative humidity in the specimen varies with the reciprocal of the saturated vapour pressure. Consequently a non-linear moisture distribution is expected.

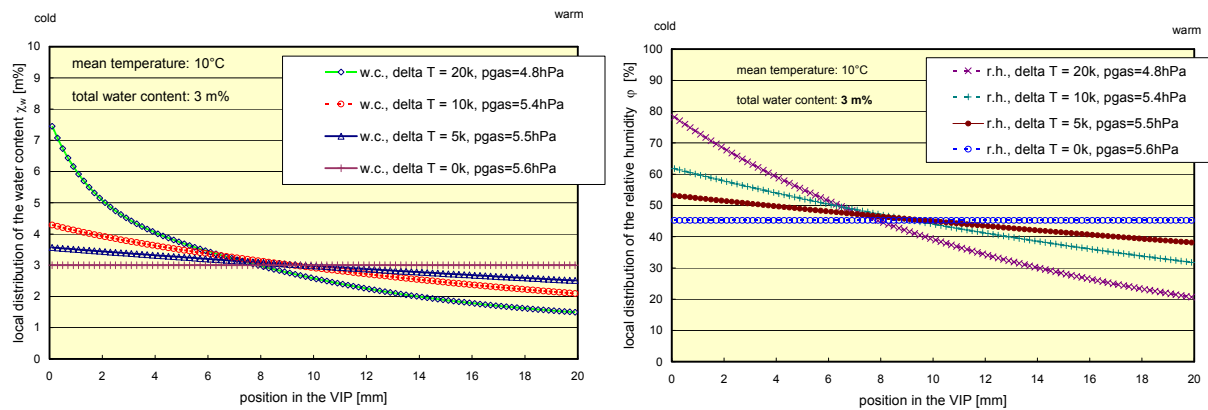


Figure 3: Moisture distribution and related relative humidity in a VIP with a thickness of 20 mm, for a total water content of 3% in mass. For the same mean temperature of 10°C, the temperature spread ΔT was varied. Averaged relative humidity is about 45%.

Numerical calculations were performed with following input parameters: the total water content in the VIP, the mean temperature and the temperature gradient. Besides material data the mean of de- and adsorption isotherms approximated as depicted in figure 2 was used, emulating the situation of a VIP that has been dried in the production process and in which water vapour has penetrated though the envelope. A typical situation of interest in practical application of VIPs in buildings is depicted in figure 3. The total water content of 3% in mass corresponds to mean humidity of about 45%. This is a humidity close to the mean annual humidity in a building façade. Two effects may become obvious from this figure:

- The non-linearity is especially pronounced when the relative humidity exceeds the linear region of the adsorption isotherm. This is the case when the temperature spread is increased to $\Delta T = 20$ K.
- The water vapour partial pressure varies with the temperature gradient. In the consequence the influence of the vapour on the total heat transfer thus also is expected to vary with the temperature gradient. Thus again in principle from the experiments an apparent thermal conductivity will be derived.

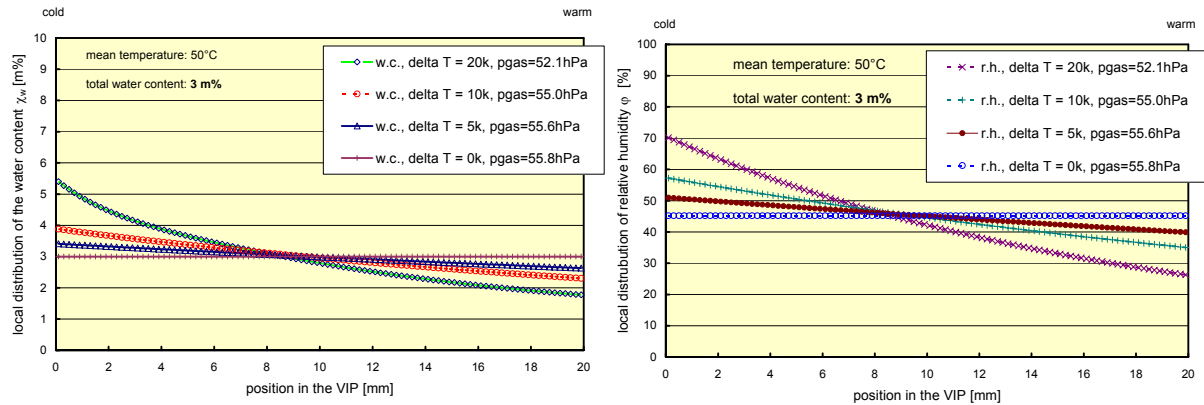


Figure 4: Moisture distribution and related relative humidity as in figure 3. Mean temperature here is 50°C.

In Figure 4 the analogous situation but for a mean temperature of 50°C is depicted. For the same temperature gradient the moisture concentration on the cold side is less pronounced at the higher temperature level.

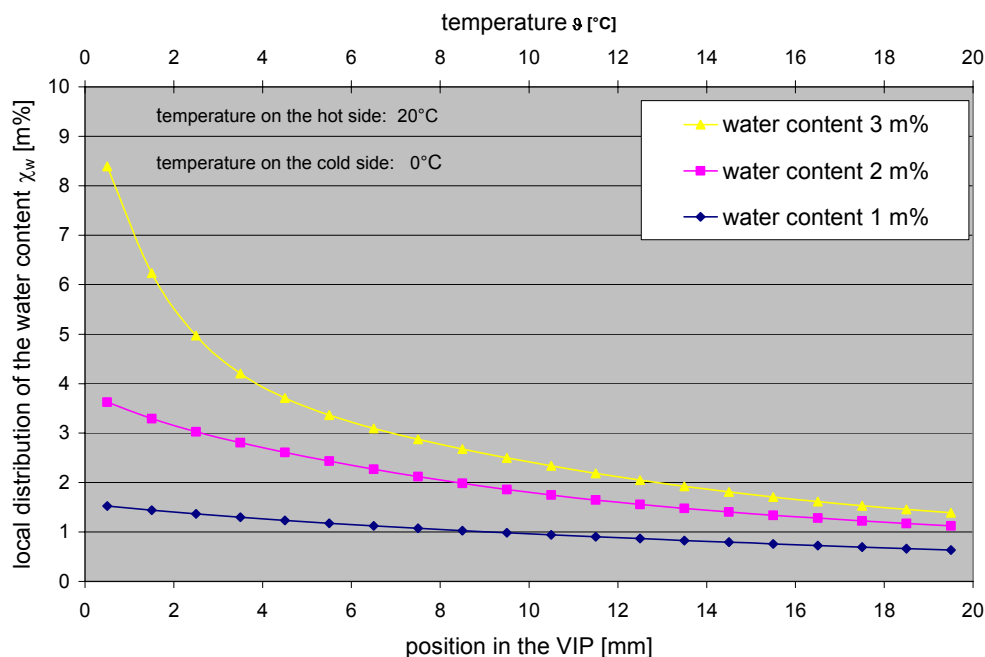


Figure 5: Moisture distribution in a VIP with a thickness of 20 mm, temperature on the hot side 20°C, on the cold side 0°C. The total water content was varied.

From figure 5 it becomes obvious that for an increasing water content most of the water is adsorbed on the cold side of the panel, if the relative humidity at the cold side exceeds the linear region of the adsorption isotherm. In this figure the adsorption isotherm was used. The approximation that no moisture back transfer along the solid backbone occurs is not realistic – except for the temperatures below 0°C, when the mobility of the H₂O-network is strongly reduced. In reality the moisture accumulation on the cold side is reduced by this back transfer and thus the shown moisture distribution in figures 3 and 4 will be flattened.

3.4 Thermal conductivity measurements with an evacuable guarded-hot-plate apparatus

3.4.1 Experimental setup

Two series of thermal conductivity measurements were performed with the ZA-constructed evacuable, pressure loadable guarded-hot-plate apparatus LOLA 4 (figure 6).

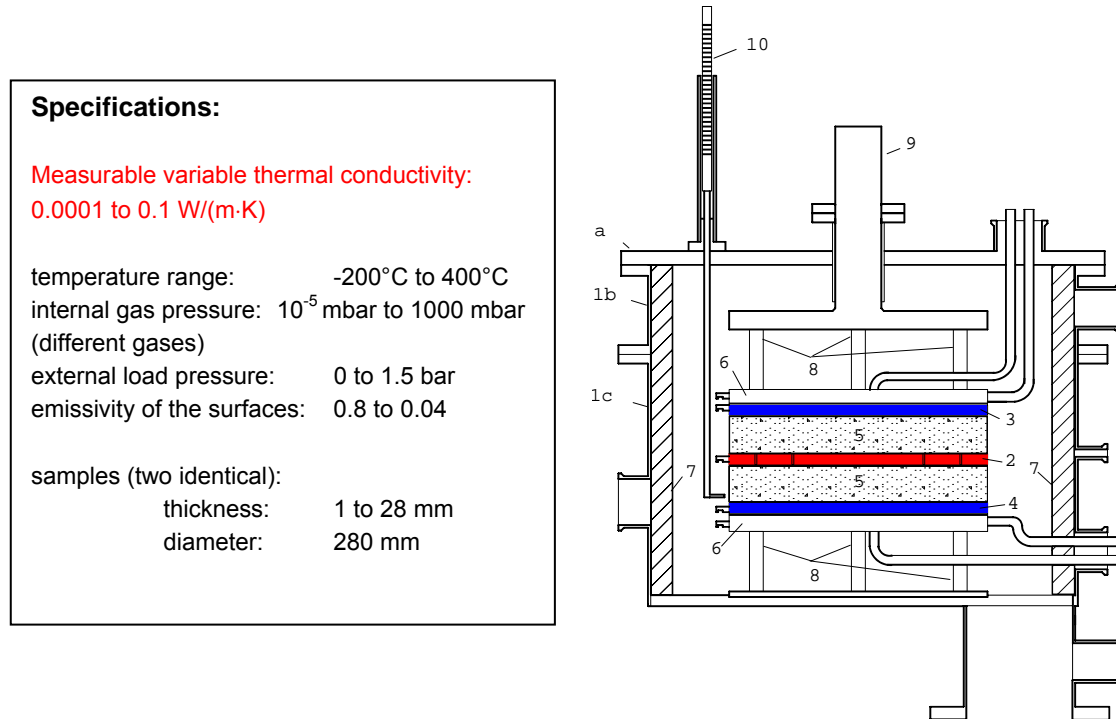


Figure 6: Diagram of the two-plate apparatus LOLA 4. 1 vacuum chamber, 2 hotplate with two guard rings, 3 and 4 cold plates, 5 samples, 6 heat sinks, 7 insulation, 8 ceramic supports, 9 vacuum-tight guided stamp, 10 three thickness sensors

The apparatus was modified in two ways. Firstly the electrically controlled cold plates were removed. The temperature on the cold sides then was directly controlled by an external fluid thermostat. The maximal achievable water vapour pressure in the apparatus is limited by the temperature of the coldest parts in the apparatus. These are either the heat sinks – for measurements at temperatures below room temperature – or the outer walls of the vacuum chamber for measurements at elevated temperatures.

Secondly a source of water vapour as drafted in figure 7 was flanged to the apparatus. It consists of a small chamber partially filled with water. Via a first valve it can be evacuated down to water vapour pressure. Via a second valve water vapour can be fed into the apparatus.

If in ‘evacuated’ moist vacuum insulations the water vapour partial pressure resembles the total pressure from the measured pressure and the mean temperature of the samples the amount of water adsorbed by the samples can be calculated.

From the variation of the temperature, the temperature gradient and the water content data for the practical applications can be gained; secondly this data should be the basis for an analytical description of the influence of water on the heat transfer in ‘evacuated’ fumed silica insulations.

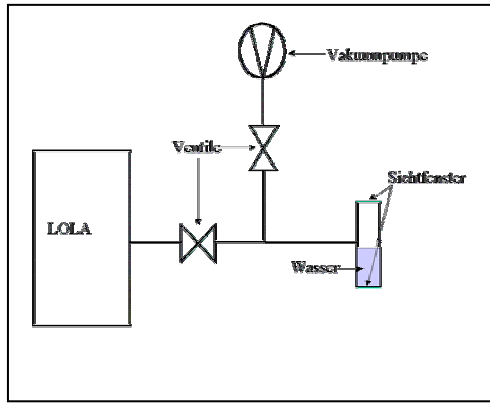


Figure 7: Diagram of the source for water vapour (not drawn to scale)

3.4.2 Measurement results

Initially the temperature and gas pressure dependencies of the thermal conductivity were determined for the dry samples (figure 8).

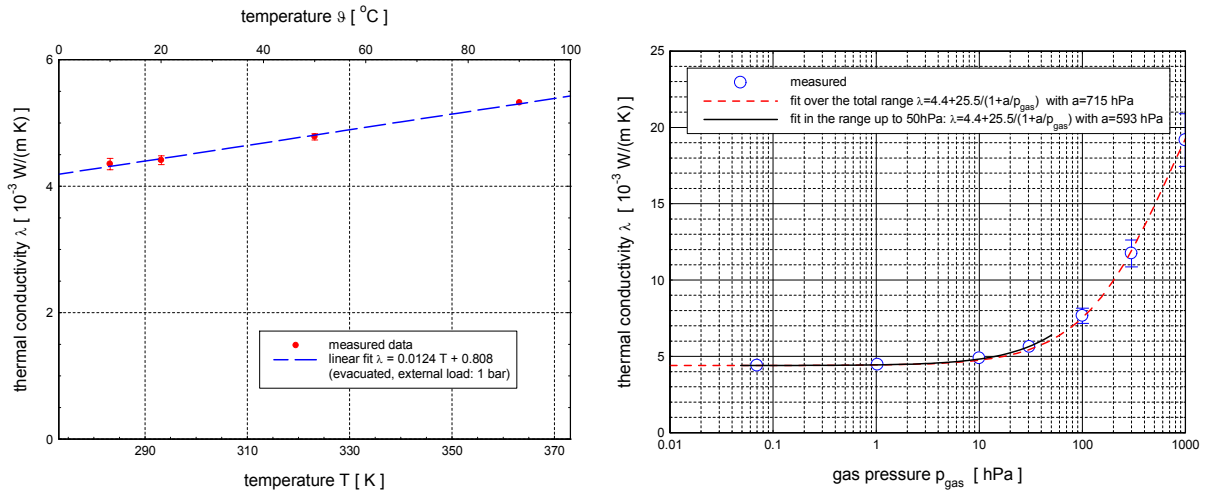


Figure 8: Thermal conductivity of the dry material versus the temperature (evacuated, left figure) and versus the N₂-gas pressure (mean temperature 20°C). For all measurements the external load on the samples was 1 bar.

Seemingly the different non-linearities in the temperature dependencies of the solid conductivity and the radiative conductivity compensate each other, thus for the temperature range investigated in a good approximation for this samples the temperature dependency of the evacuated specimen can be described by a linear function:

$$\lambda_{\text{evac}} / 10^{-3} \text{ W/(m K)} = 0.0124 \cdot T / \text{K} + 0.808 \quad \text{Equation (4)}$$

From the gas pressure dependency a characteristic pressure $p_{1/2} = 715 \text{ hPa}$ is derived when performing a fit for the total range up to 1 bar. A somewhat smaller value of about 600 hPa is found, when applying a fit only up to 50 hPa, the range where the influence of water vapour pressure will be studied in detail.

In figure 9 the results of several sets with different amount of water in the apparatus are depicted. From the left picture in figure 9 linear dependencies of the conductivity on the water content seems to hold for all temperatures investigated. As one might expect from the increase of water vapour

pressure with temperature for a fixed humidity (vertical line) the effect of moisture also increases with temperature, resulting in an increased slope in this figure for higher temperatures.

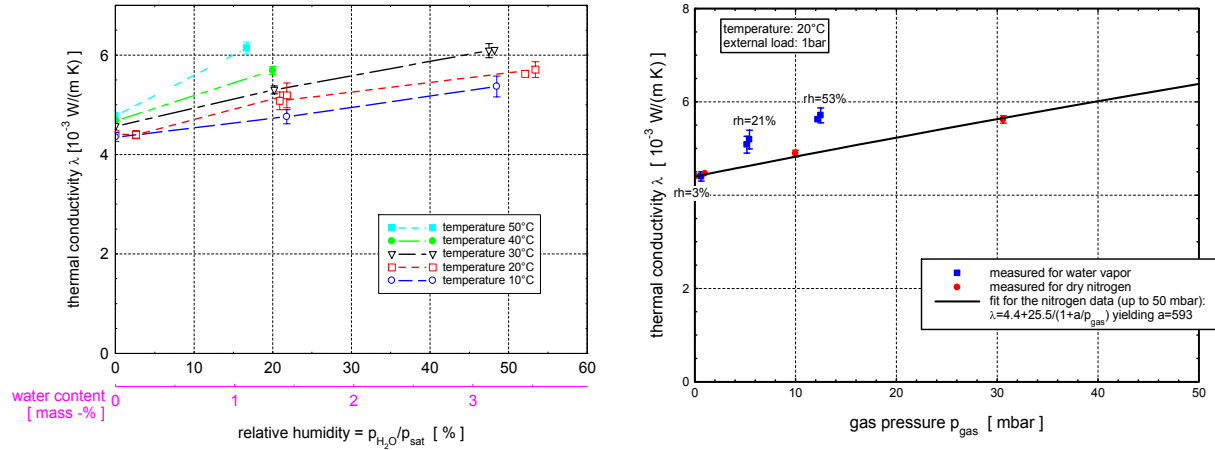


Figure 9: Measured thermal conductivity as a function of different relative humidity for different mean temperatures (left), and values obtained for 20°C for different H₂O and N₂-gas pressures (right). The temperature spread always was 20 K.

The right picture of figure 9 resembles conditions close to the situation in building applications (mean temperature 20°). The measured thermal conductivity is depicted for different water content versus the corresponding H₂O-gas pressure and the N₂-gas respectively. Both variations may be interpreted as the maximum degradation effects for vacuum insulation panels employed in buildings: the onset in the conductivity by penetrating air (assuming a pressure increase by air of 1 mbar per year) and in the extreme case an equilibrium of internal and mean annual humidity outside a VIP.

The second series of measurements was thought to improve the data basis for theoretical considerations, especially to check, whether the conductivity increases caused by air and by moisture may be simply summed up.

Even though the relationship of the total heat transfer and moisture content, temperature and temperature gradient may be very complex, a simple approach was tested to describe the total thermal conductivity as function of humidity and temperature.

Some assumptions and approximations were included:

- At first only measured data have been selected, where the temperature spread was 20 K.
- The temperature dependence of the conductivity of the evacuated, dry specimen was assumed to be linear, as found above.
- For the low gas pressure range considered here, the Knudsen formula was approximated as

$$\lambda_{\text{gas}} = \lambda_{\text{gas},0} / (1 + p_{1/2} / p) \approx \lambda_{\text{gas},0} / p_{1/2} \cdot p = \frac{\lambda_{\text{gas},0}}{p_{1/2}} \cdot p, \quad \text{Equation (5)}$$

- As temperature effects of the conductivity of free, still gas $\lambda_{\text{gas},0} \propto \sqrt{T}$ and that of the mean free path $l \propto T$ are opposed, the resulting effect is expected to be small for the small temperature range considered here. Thus no temperature dependence was assumed for the ratio $\lambda_{\text{gas},0}/p_{1/2}$.
- For the adaption of the different kind of gases (H₂O and N₂) in the Knudsen formula for the same reason a constant factor was used.

The most influencing variables are assumed to be:

- adsorbed water in the 'liquid' phase, which is assumed to increase the 'solid' conduction proportional to the amount of adsorbed water, and
- in the gaseous phase by the partial water vapour pressure $p_{H_2O}(T) = \varphi \cdot p_{sat}(T)$ with

$$p_{sat}(T)/hPa = 6.11 \cdot \exp\left(17.08 \frac{T/K - 273.15}{T/K - 39}\right), \text{ the analytical description of the saturated water vapour pressure.}$$

From this the fit formula results:

$$\lambda = \lambda_{vac}(T) + c' \cdot X_w(\varphi) + d \frac{\lambda_{gas,0}}{p_{1/2}} \cdot p_{H_2O}(T) \quad . \quad \text{Equation (6)}$$

With the values given above and use of a linear adsorption isotherm one gets:

$$\lambda = (a + b \cdot T) + c \cdot \varphi + d \frac{25.5}{600} \cdot p_{H_2O}(T) \quad \text{Equation (6)}$$

A 4-parameter fit for a , b , c and d with measured λ , φ and T yields a satisfying agreement between measured and calculated data for all parameters varied:

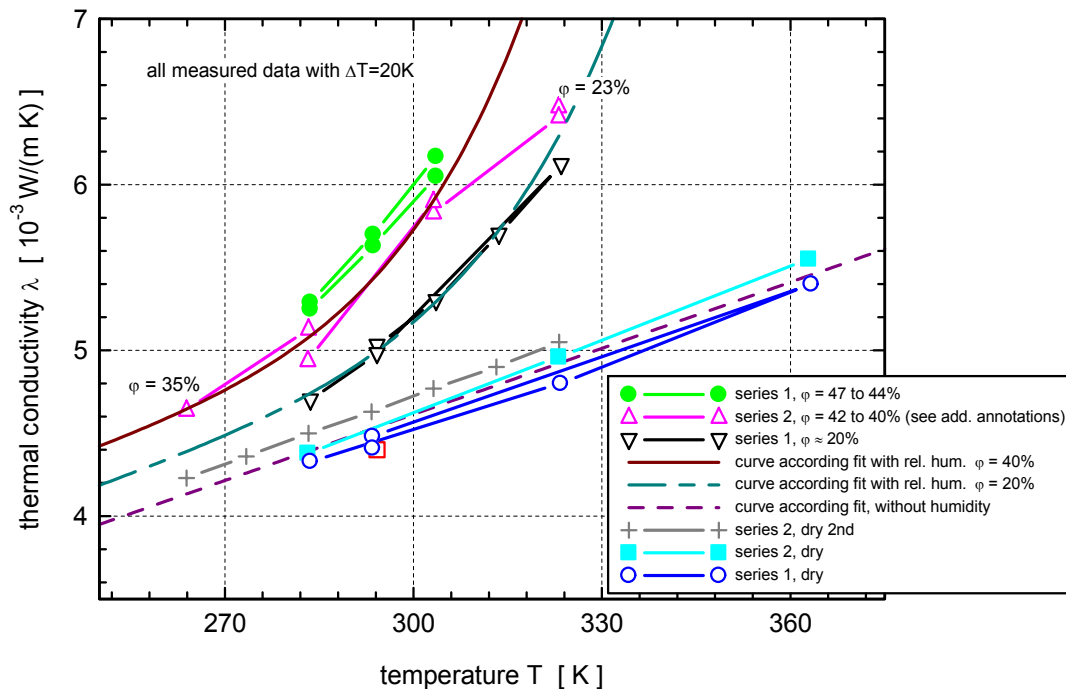


Figure 10: Measured thermal conductivity for different water content and temperature. Additionally from the 4-parameter-fit calculated conductivity of the evacuated specimen is depicted, as well as for specimen with 20% and 40% relative humidity.

The resulting fit parameters are:

$$a = 0.624 \cdot 10^{-3} \text{ W/(m K)}, \quad b = 0.0133 \text{ /K } 10^{-3} \text{ W/(m K)}, \quad c = 1.14 \cdot 10^{-3} \text{ W/(m K)} \quad \text{and} \quad d = 1.08.$$

In figure 10 the measured data are compiled together with curves calculated according to the fit result, i.e. the conductivity of the evacuated specimen versus temperature and the conductivity of specimen with an relative humidity of 20 and 40%.

It has to be pointed out, that in the course of the different series of measurements with different moisturisation and drying cycles, the thickness of the samples shrank from 18.6 mm to 18.2 mm. Parallel the conductivity of the evacuated specimen was increased by about $0.4 \cdot 10^{-3} \text{ W/(m K)}$, as may be seen from the three lowest curves in figure 10. It has to be checked, whether possibly capillary forces at high humidity may be the reason.

In the two dimensional graphic of figure 10, different sets of measurements are assembled to graphs for distinguished relative humidity which in the experiments was not exactly constant. Having this in mind the calculated curves for 20% and 40% relative humidity coincide quite well the measured conductivity for water contents quite close.

From the fit parameter c an effect of adsorbed water on the thermal conductivity of about $0.01 \cdot 10^{-3} \text{ W/(m K)}$ per % of relative humidity results.

The result for the fit parameter d close to 1 indicates that the onset of gaseous contributions to the total thermal conductivity is quite similar for water and for nitrogen.

4 Conclusion

The thermal conductivity of an 'evacuated' fumed silica board has been measured as a function of moisture content and temperature. Neglecting possible effects of mass transfer by vapour diffusion and counter surface and capillary diffusion a simple model describes the influence of the moisture on the total conductivity as an increase in the solid conductivity proportional to the amount of the adsorbed water and an effect of the vapour that is quite close to that of nitrogen gas. In the low pressure region, typical for practical applications of VIPs, the increase caused by the water vapour again is proportional to partial pressure.

Based on the adsorption isotherm numerical calculations describe the moisture distribution in a VIP for a broad range of water contents, temperatures and temperature gradients. Parameter studies indicate that in the measurement practice a dependency of the heat transfer coefficient on the temperature gradient is to be expected.

A more detailed analysis of the data also with different temperature gradients should give more insight into the moisture distribution and its effect on the total (apparent) thermal conductivity.

Up to now the question whether heat pipe effects also have an impact on the total heat transfer could not be answered.

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