Thermal insulation testing concept for a high temperature furnace under different atmospheres and pressures

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The thermal conductivity of porous fire-resistent and insulating materials depends on the type of solid material, temperature, the pores filling gases and the pressure. In general the manufacturers are stating the thermal conductivity of materials only for air under normal pressure. The measured values for other gases and pressures are hardly available. A reliable method for the conversion of measured values to other gases and pressures is not yet known.

For the manufacturers of furnaces this means that thermal losses in the walls of the furnaces for special gases, high pressures and vacuum could only be estimated with a high uncertainty. The mentioned problem caused under- and overdimensional heaters or insulations.

For the solution of this problem serves an electrical resistent-heated high temperature furnace „Rubistar 100" manufactured by the company Linn High Therm GmbH. This furnace is equipped with a three layer fibre insulation for a maximum temperature of 1800 °C under air and protective gas atmosphere, with a working pressure up to 100 bar. For technically interesting combinations of temperature, pressure and gas atmosphere the required heating power is determined. With these measured values an extrapolation for higher temperatures and pressures is done. The calculated specific power values allow an economical power dimensioning of other sizes of furnaces. Finally the temperature homogeneity inside the furnace chamber is measured to clarify if such types of furnaces can meet the requirements of the customers.

Introduction

For dimensioning of high effective insulation systems and for a cost estimation an exact knowledge of the thermal conductivity of the insulation material is necessary. In most cases the available, measured data do not cover suitably to the interesting field of application. This is especially true if an insulation under vacuum should be realised, because hereby the gas pressure serves as an additional parameter next to the characteristics of the material and the temperature. Also the operation under over pressure is an interesting application. From the literature [1, 2] a number of investigations is known where model equations for calculating the effective thermal conductivity are derived with simplified physical conditions.

Because of the amorphous heterogeneous structure of most insulation materials each model requires the introduction of constants in order to adjust to the data which has been found in experiments. For doing comparison and optimization calculations it is necessary that at least small groups of insulation materials are described through an unified set of adaptable constants. Hereby especially the influence of the raw density and the particle size or the fibre thickness has to be considered in a quantitative and correct way.

The models in literature which have been checked regarding the experimental data do not fulfill the requirements. Although they allow a description of the dependence of the effective thermal conductivity of pressure, gas type and temperature, but not of the insulation material itself.

The aim is therefore to check and improve the knowledge of the influence of gas pressure and as gas composition as well as the temperature and the type of material.

Thermal transport mechanisms in porous materials

Heat insulation materials are heterogeneous porous multi phase bodies. For modelling the thermal transport in such bodies the analogue laws of electrical and thermal conductivity are often used. The relation between characteristic length l_k to the product of thermal conductivity λ and flowing cross section A is called thermal resistance R, so that

$$\dot{Q} = \frac{\Delta T}{R} \qquad (1)$$

analogue to the Ohm's law with the heat current $\dot{Q}$. Analytical models for the description of the thermal conductivity in porous bodies are mostly build with the help of parallel and serial circuits of thermal resistances. To the single components of the structure thermal resistances are assigned according to the conditions of the thermal transport. Depending on the modelling of the different connections this results in mathematical equations which contain the thermal conductivity as a function of porosity, more structure characteristics, temperature and the thermal conductivity of the solid and gas phase. Fig. 1 shows an example of possible connection of the resistances, which contains all well-known thermal transport processes.
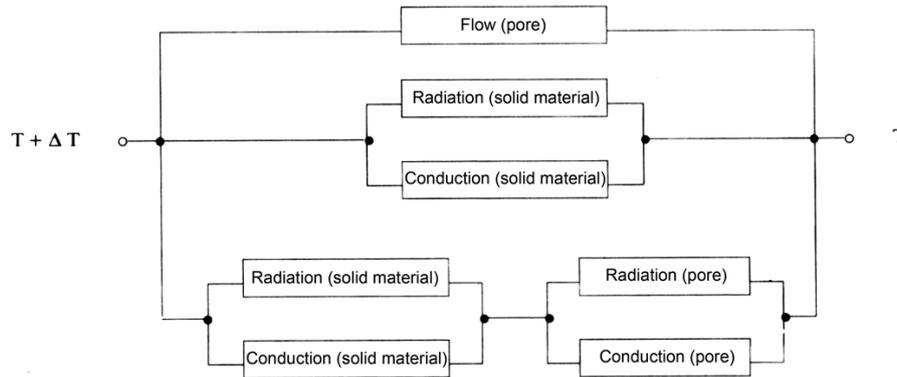
Fig. 1 Model scheme of thermal transport in multi phase, porous and heterogeneous materials, shown in a network of resistances [1]

So thermal energy can be transported via four ways from the position with the temperature T + ΔT to the position with the lower temperature T:
- By a flow through the pores, where the flowing medium transports the energy
- By conduction and/or radiation of the pore-free solid material
- By conduction and/or radiation of the solid material interrupted by the pores

The model shown in fig. 1 can be displayed a lot more simplified for different insulation materials. In common materials e.g. in fireclay bricks stone the thermal transport is effected through thermal conductivity in the solid material and radiation inside the pores. In fig. 1 this special case only contains the resistances conduction (solid material) in the middle path and the resistances conduction (solid material) and radiation (pores) in the lower path. On the other hand for each fibre insulation a large thermal quantity can be transported due to a flow through the fibres with their low density and by the radiation inside the amorphous fibres which makes the analytical description much more difficult.
The standard experimental setups for calculation of the thermal conductivity do not detect the thermal transport caused by gas flow through the insulation materials. However in practical use this kind of transport occurs.
The general case of the behaviour of the thermal conductivity λ using a mixture of two phases with different is shown in fig. 2 where the model in [2] is applied. The relation between λ is used as parameter in the curves. So the porosity reduces the thermal conductivity quite a lot, if the gas phase is continuous. If the phase is distributed discontinuously inside the pores, the thermal conductivity decreases approximately linear with the porosity. This was confirmed according to experiments with fireclay bricks and alumina refractory up to 60 % porosity for temperatures up to 1500 °C.
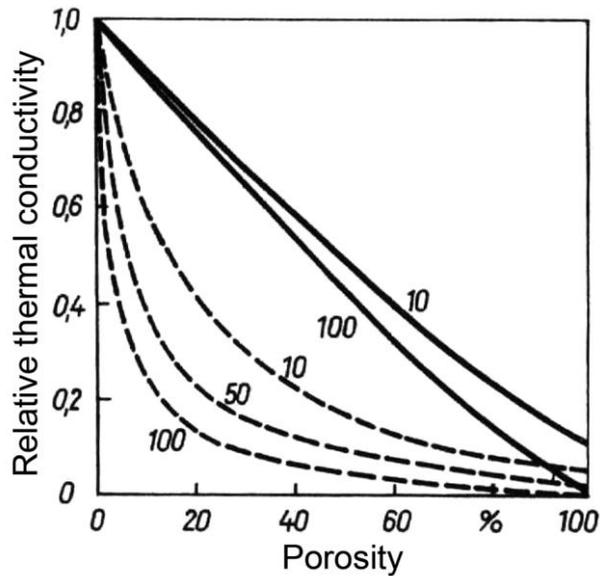
Fig. 2 Relative thermal conductivity for different mixture relations of two phases with different thermal conductivity. This relation is plotted as function of the porosity [1]

For increasing porosity the processes mentioned in fig. 3 are deviating from the models which were mentioned before. In fig. 3 the dashed lines are showing different thermal transport processes. When the porosity increases also the relative quantity of gas inside the insulation material is increasing and the thermal transport in the gas phase will grow as well, equivalently the thermal conductivity will be reduced due to the solid phase. The components of the thermal flows sum to the effective thermal conductivity due to different transport mechanisms, which are shown in fig. 3 with the full lines. The effective thermal conductivity always has a minimum depending on the porosity, if radiation or convection participates in thermal transport. The minimum is shifted to lower porosities with increasing temperatures, because the radiation increases more with higher temperatures than the conductivity. At porosities under the minimum of the thermal conductivity the effective thermal conductivity becomes very similar to the shape in fig. 2. The occurrence of a minimum thermal conductivity in dependence of the porosity means that there exists an optimal porosity and raw density, where the thermal insulation has a maximum.
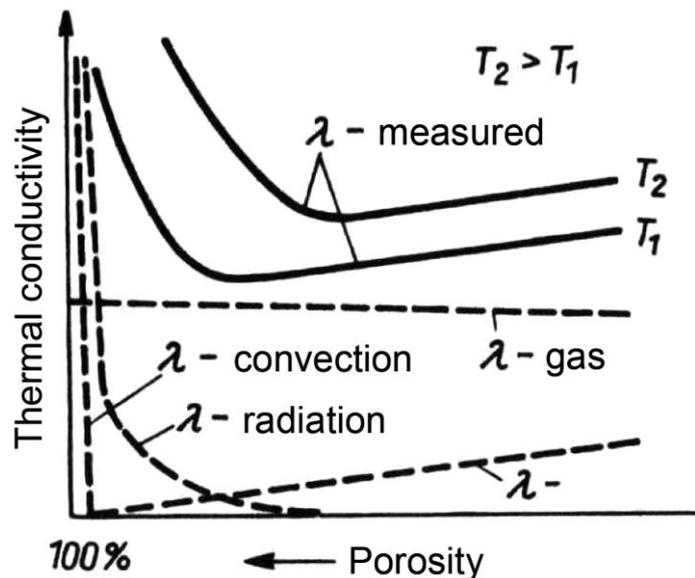
Fig. 3 Thermal transport mechanisms (dashed lines) and effective thermal conductivity (full lines) concerning high porosities [1]

The thermal conductivity is roughly linearly dependent on the pore size as shown in different models. When the pore size is smaller than the average free length of the gas molecules (smaller than 100 nm), a big reduction of the thermal conductivity of the pore gas can be observed. So it is explained that the so called microporous insulation materials have a lower thermal conductivity than static air.

Definitions regarding the effective thermal conductivity

The effective thermal conductivity of an insulation material is the result of the elementary thermal transport processes. The differential equation

$$\frac{d}{dA}\left(\frac{\partial Q}{\partial t}\right) = -\lambda \cdot grad\,T \tag{2}$$

is not only applied for homogeneous, but also for heterogeneous multi phase and porous materials, where is: grad T = local temperature gradient, Q = thermal energy, λ = thermal conductivity, t = time, $\frac{d}{dA}\left(\frac{\partial Q}{\partial t}\right)$ = differential heat flow through the area element dA.

The thermal conductivity of an insulation material measured in experiments and given in tables is not only a value for thermal transport by conduction anymore, however it is valid for the complete thermal transport not depending on the mechanism inside the material. This effective thermal conductivity includes the complete thermal transport caused by conduction, radiation and eventually convection. The effective thermal conductivity is a material property, which gives an information about the heat flow inside a temperature field, which flows due to the temperature difference over a defined length across an area in direction to the normal axis.

The thermal conductivity of insulation materials increases with the temperature, if it is lower than 1 W / (m · K) at room temperature, and it decreases, if it is much higher than 1 W / (m · K) at room temperature. This behaviour can be explained by the different portions of pores and solid material and the radiation transport in highly porous materials (very low thermal conductivity at room temperature, however high thermal conductivity at high temperature). In contrast to this, low porous materials have a high thermal conductivity because of the high thermal conductivity at room temperature of the solid phase, at high temperatures the thermal conductivity is lower because of the negative temperature coefficient of these solid materials.

For porous multi phase materials the empiric relation with the effective thermal conductivity λ_{eff} can be written as [1]:

$$\lambda_{eff} = a \cdot \frac{1}{T} + b \cdot \sqrt{T} + c \cdot T^3 \tag{3}$$

The first summand in equation (3) describes the pure thermal conduction of the pore free crystalline phase. There the constant a describes how the thermal conductivity depends on the material. The second summand gives information about the pure thermal conduction inside the pores and in a non-crystalline material. The constant b should be inverse proportional to the total porosity. The third summand is responsible for the inner radiation, whereas the quantity constant c is a measure the heat transport by radiation. So for insulation materials the following facts can be derived:

- The share of the inner radiation increases much more with higher temperatures than the conduction portion. At high temperatures the transport takes place mainly by inner radiation.
- The share of conduction decreases with increasing total porosity at constant temperature.
- The share of radiation decreases with smaller average pore size at constant temperature. It strongly depends on the distance between the radiation areas and so on the pore size distribution.
- An insulation material with a high empty space part and smaller average pore size will show a lower increase of the effective thermal conductivity depending on the temperature
- The effective thermal conductivity can depend on the direction, in the case of anisometry or textures, especially at random pore volume structures.

When all facts are summarized, there are the following requirements for an insulation material with minimum thermal conductivity.

- The porosity has to be adjusted to the working temperature and the temperature distribution. The thermal conductivity decreases with increasing temperature.

- The pore size should be as small as possible.
- Cracks and rough pores, which allow a convective thermal transport, have to be avoided. This has to be taken care at a sequence of several insulation materials.

A material with maximum thermal conductivity can be reached in the following way:
- The material must be free of pores.
- The solid phase should consist of a crystal structure with high density, where atoms with very low relative mass are used. Furthermore the crystal size and the regularity should be very high (no micro cracks or amorphous parts). The material must consist of a single phase and have a good transparency.
- Additional components like graphite and silicon carbide (SiC) improve the thermal conductivity of the most refractory materials. For special applications BeO and AlN are the best choice with the highest thermal conductivity.

Influence of the pressure on the thermal conductivity

Experiments have shown that the thermal conductivity of a gas inside a porous medium can differ from the pure gas value. So dimensioning of the including space, e. g. pores in insulation materials, is an important factor. This phenomenon can be explained by the kinetic gas theory, where the Knudsen number Kn describes the relation between the average free length of the gas molecules l_g and the pore size d_p.

$$Kn = \frac{l_g}{d_p}$$ (4)

According to the kinetic gas theory the thermal conductivity λ_{g0} of an ideal free gas is calculated (c_g: spezific heat capacity, ρ_g: density, v_g: average velocity of the gas molecules, l_d: average free length of the gas):

$$\lambda_{g_0} = \frac{1}{3} \cdot c_g \cdot \rho_g \cdot v_g \cdot l_d$$ (5)

For more details see [3].
Three cases can be distinguished:
- $l_g \ll d_p$, the thermal conductivity λ does not depend on pressure
- $l_g \gg d_p$, the thermal conductivity λ is proportional to the gas pressure at constant temperature, at constant pressure λ is proportional to $1/\sqrt{T}$
- $l_g \cong d_p$, the thermal conductivity does not depend on pressure, more detailed facts cannot be derived.

The first case happens, if the pore size or the distance between the surrounding walls d_p is constant and much bigger than the average free length l_g of the gas molecules. The second case will appear if the pores are too small and they have the same size. However this cannot be observed in reality because most pores have different shapes and diameters.
This is shown in the fig. 4 (left): At constant pore size this value can be compared with the average free length of the gases. The Knudsen number is 1 or larger than 1 at low pressures. The average free length decreases by increasing the pressure [3], which causes a decreasing Knudsen number. So the thermal conductivity does not depend on the pressure.
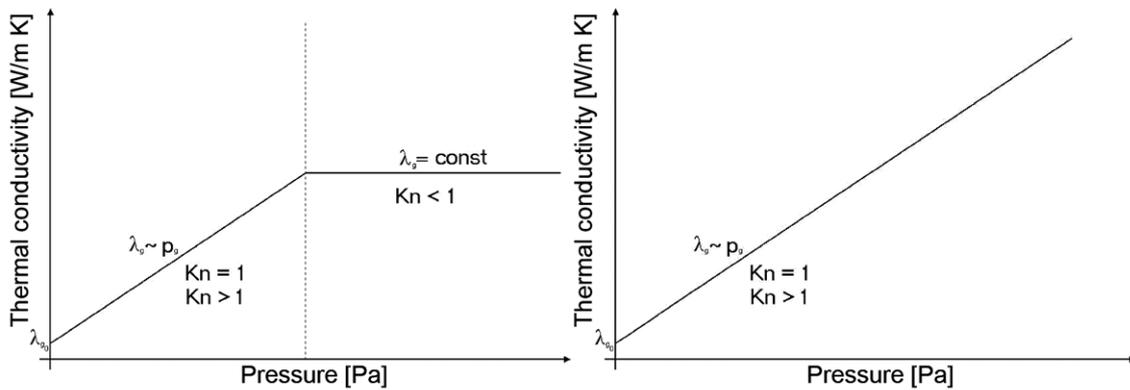
Fig. 4 Dependence of the thermal conductivity at constant (left) and different (right) pore size

For different pore sizes (fig. 4 right) the size of the micro pores is very important, these pores, where the size is much smaller than the average free length of the gas molecules, have an influence on the thermal conductivity of the insulation material.

During verification of this theory in experiments there appears the difficulty, that in general thermal process units the thermal conductivity of the insulation materials cannot be calculated so easily. However there is a direct relation to the required heating power, which has to be installed by the furnace builder, in order to fulfil customer's requirements of heat up time and temperature accuracy. Power can be measured easily, and tendencies can be transferred directly to the effective thermal conductivity. So the measurement results can be compared with the theory. That's why in all experiments only the required heating power is measured.

Experimental set-up

For the experiments a high pressure furnace type "Rubistar 100" of the company Linn High Therm GmbH is used (fig. 5). This furnace is applied for heat treatment of corundum (rubies, sapphires) using the diffusion method (fig. 5), in order to improve the colour, or for sintering of powder metals (used for hard metal cutting devices).

Fig. 5 High pressure furnace "Rubistar 100" of the company Linn High Therm GmbH, right above treated corundes like rubies and sapphires (Corundum Mine Co. Ltd. Bangkok), below hard metal cutting devices (Boehlerit)

The furnace has a box shaped useful volume with the dimensions 70 mm x 80 mm x 120 mm (w x h x d). In these experiments a three sheet insulation (combination of fibre and microporous material) is used and allows a short time temperature of max. 1800 °C, 1750 °C continuous temperature under oxidizing atmosphere The highly effective insulation promises a temperature accuracy of ± 10 K, the high pressure vessel allows pressures up to 100 bar under air, argon and nitrogen (or mixtures of these gases with hydrogen [max. 5 % H_2]). Before starting the process the chamber is evacuated down to $5 \cdot 10^{-3}$ mbar by a vacuum pump. Kanthal Super 1900 heating elements with a typical power of 5 kW are installed.

The double walled vacuum chamber consists of the furnace body and two lids. Although there is used a highly effective insulation, the chamber has to be water cooled, in order to reach a maximum surface temperature less than 60 °C. Simple fresh water is sufficient for this. The machine has two connectors for protective gas and air, the last one also can be used for a second protective gas. The front side of the chamber has a square opening for charging, at the top side the lead throughs for the heating elements are installed (fig. 6 left).
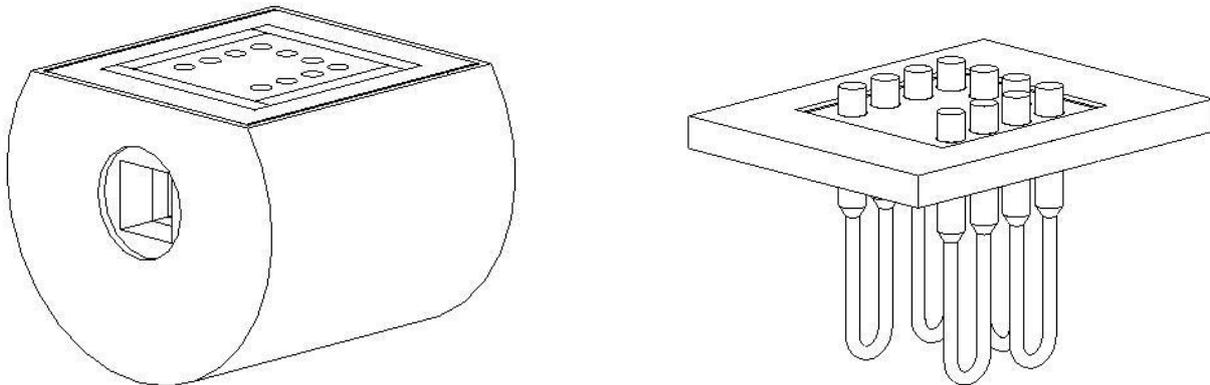
Fig. 6 Insulation set (left) and Kanthal Super 1900 heating elements (right)

The chamber is equipped with five Kanthal heating elements, which are installed vertically and connected with a aluminium rail at the high pressure current lead through (fig. 6 right).

A maximum temperature of 1800 °C in combination with the applied protective gases is allowed. Only when using hydrogen (5 %) the maximum temperature decreases down to 1650 °C. As in every resistance heated furnace, the chamber temperature is 30 to 100 K lower than the heating element temperature.

Four thermocouples are used for temperature measurement, two are responsible for measurement inside the furnace chamber (Pt40Rh/Pt20Rh). This first one is connected to the safety controller, the second one works together with the standard controller.

The third thermocouple (type S Pt-Rh), which is installed inside the center insulation layer, detects temperature changes during heat up. The fourth thermo couple (type K NiCrNi) is integrated at the inner surface of the pressure vessel and it shows changes of the inner wall temperature. Fig. 7 shows the complete thermocouple setup.
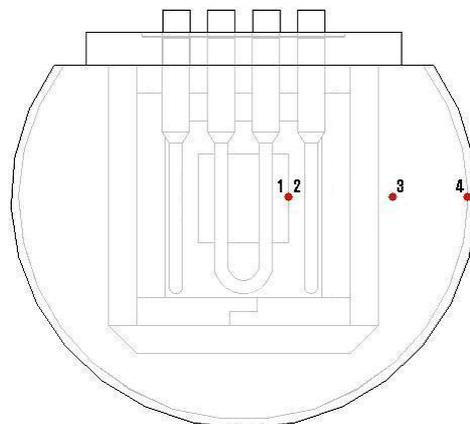
Fig. 7 Setup of the thermocouples in front view into the furnace chamber

Practical experiments

The following program was the basis of the experiments:
- Heating up as fast as possible starting from the cold state to the final temperature of 1000 °C or 1700 °C
- Keeping this temperature constant over three hours
- Cooling down to 300 °C with water flow while decreasing the pressure
- Opening the furnace at less than 300 °C

All experiments were done under the following atmospheres with the following pressures:
- Vacuum better than $5 \cdot 10^{-3}$ mbar
- Air: normal pressure, 10 bar, 25 bar, 50 bar, 75 bar and 100 bar
- Argon: normal pressure, 10 bar, 25 bar, 50 bar, 75 bar and 100 bar

The following temperatures were measured:
- Chamber (thermocouple 2)
- Center of insulation (thermocouple 3)
- Inner surface of pressure vessel (thermocouple 4)

Furthermore the following values were measured:
- Vacuum pressure (only in experiments under vacuum)
- Heating power
- Cooling water flow
- Cooling water inlet and outlet temperature

In the next step the temperature accuracy inside the chamber was investigated using ceramic rings (diameter app. 25 mm). These rings consist of ceramic materials and they are available for different temperature ranges from nearly 1000 °C up to app. 1800 °C. The principle is used that the rings shrink depending on the achieved temperature. After measuring the final diameter, the corresponding temperature can be read in the tables of the rings' manufacturer. However the rings should lie on fine aluminium oxide powder in order to reduce friction which can falsify the measurement. Furthermore such an experiment is only reliable, when the temperature was kept constant for at least three hours. Fig. 8 shows the arrangement inside the furnace chamber, all rings were fixed at the half height. So an investigation in one only horizontal layer was done.
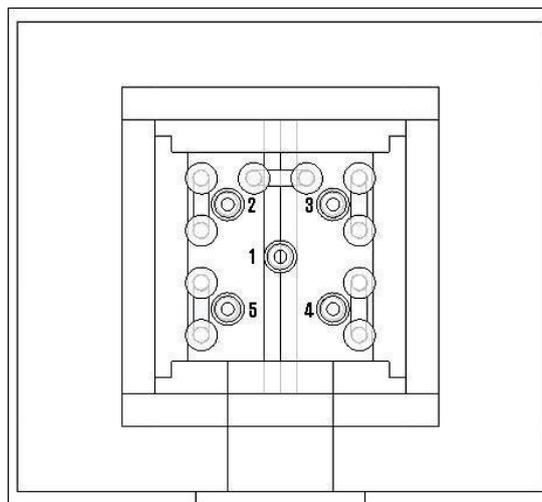
Fig 8 Arrangement of the ceramic rings inside the furnace chamber (top view). Left, right and at the back side the heaters are visible.

Dependence of the heating capacity from pressure and temperature

For every combination of gas, pressure and temperature the heating curve is plotted. Because the furnace requires much time for reaching the final temperature (heating power around 5 kW), the theoretical heating power is calculated by extrapolation. Fig. 9 shows the heating power and the required wa-

ter cooling power depending on the time (example for 100 bar argon with 1000 °C final temperature). After four hours the measurement was stopped and the power values for the stationary state were calculated by extrapolation. The cooling water power losses can be calculated by the temperature difference between inlet and outlet and the constant water flow.
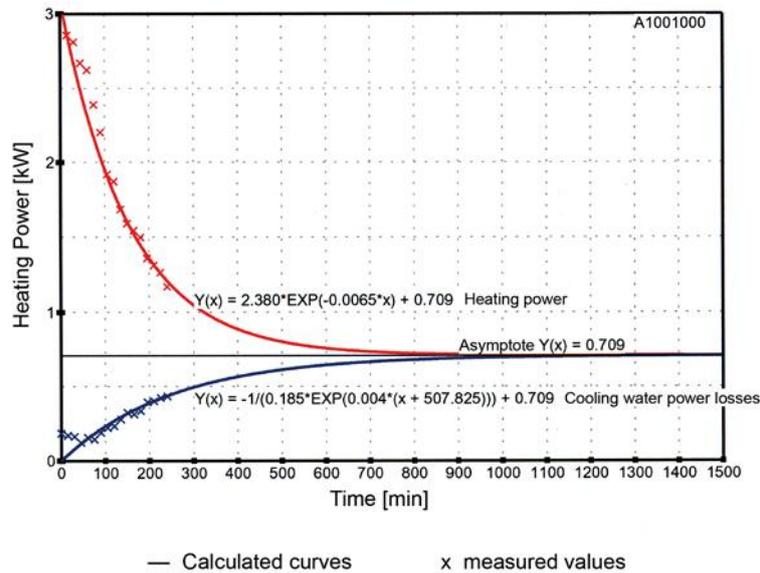
Fig. 9 Measurement curve for 1000 °C final temperature under 100 bar argon atmosphere

The asymptotic value of the heating power is not equal to the real value, which has to be installed by the furnace manufacturer, however the user receives relative heating power values depending on atmosphere, pressure and final temperature.
The detected asymptotic heating power values for the stationary case are plotted as function of the pressure (fig. 10).
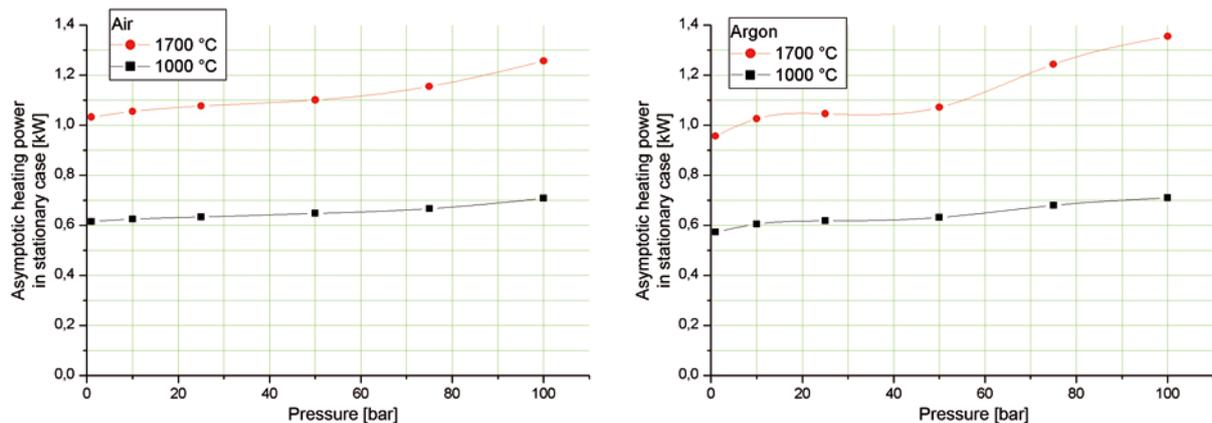
Fig. 10 The stationary heating powers calculated by extrapolation depending on pressure for air (left) and argon (right)

As the results of air and argon do not differ strongly, the following analysis is only done for argon, here the required power is maximum 10 % more than for air.
The investigations were only done for the temperatures 1000 °C and 1700 °C. However the product range of Linn High Therm goes up to 2100 °C. Higher temperatures would only be feasible with other kinds of insulation materials. Because the thermal conductivity differs from those which were used her, the results cannot be transferred.
With constant pressure for the interpolation between 1000 °C and 1700 °C and for the extrapolation to 2100 °C approximately a polynomial function of third degree can be assumed. For every pressure three measuring points are taken (0 kW for room temperature and the measured values for 1000 °C and 1700 °C) for the interpolation with a polynomial of third degree.
In order to transfer the results to other furnace chamber sizes it is useful to use the specific power, the

power per liter useful volume, instead of the absolute power. In the last step finally the conversion of the stationary power to the real installed power is done. However this is not easy to do only using the measured data, here the long experience in furnace building is a very big advantage. As reference the fact is used, that a specific power between app. 12 and 15 kW is required, if 2100 °C maximum temperature has to be reached within two hours with a pressure of 100 bar. Fig. 11 shows the finally calculated curve group.
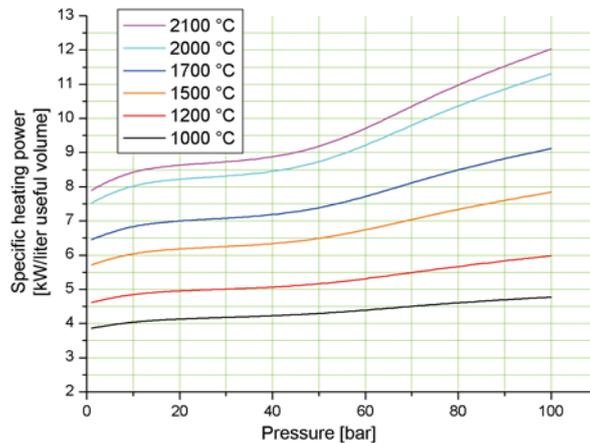
Fig. 11 Curve group for dimensioning of high temperature furnaces at pressures up to 100 bar under air and argon atmosphere

These curves only may be transferred to high pressure furnaces of the same type, this means the thickness of all insulation layers and the type of material may not differ essentially in the thermal conductivity (also depending on the temperature).
The shape of the curves can be explained by the dependence of the thermal conductivity on the pressure. Obviously the applied insulation materials have approximately a constant pore size, so the curve in fig. 4 can be transferred directly to the above mentioned curve group. First the thermal conductivity increases (Kn $\geq$ 1), so the required power also increases in the same way, this corresponds to the range up to 80 bar. At higher pressures the Knudsen number decreases to 1, that's why up to 100 bar a lower increase of the curve group is observed.
More values were measured in vacuum operation, the same calculation of the specific heating power and the same calibration were done as in the high pressure operation. As result can be mentioned that the required power is only 2/3 of the value at normal pressure mode (1 bar). Fig. 12 shows the comparison of the specific power between vacuum mode and normal pressure mode for the temperature range from 1000 °C to 1700 °C. This behaviour can also be explained by the linear increase of the Knudsen number at lower pressures – a reduction of the pressure down to 10^{-3} mbar causes a reduction of the thermal conductivity in accordance to fig. 4. Also in literature [6] is mentioned that the thermal conductivity in vacuum is only app. 70 % of the value at normal pressure.
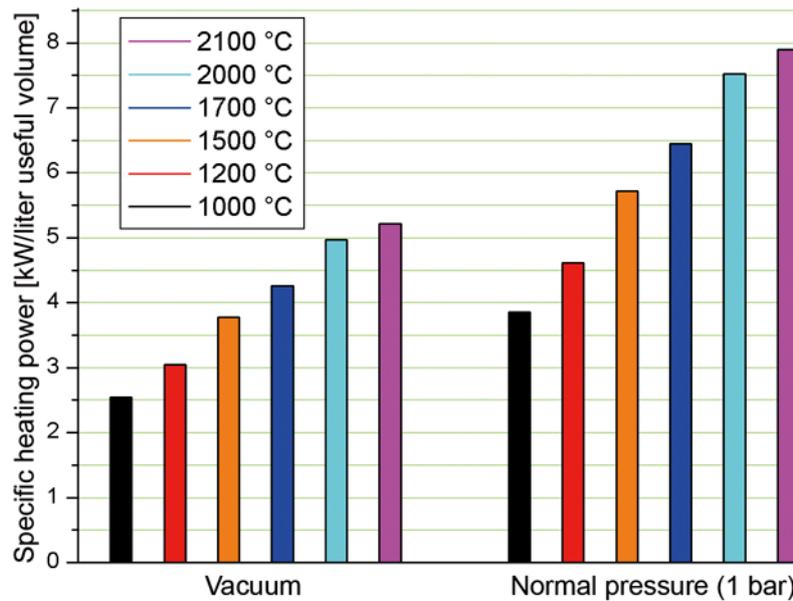
Fig. 12 Rough specific power values for dimensioning of high temperature furnace under vacuum

Temperature Uniformity

In air the temperature uniformity was investigated by using ceramic rings, in order to detect the influence of the pressure on the homogeneity. This is a very important fact, because there is an increasing market demand for an uniformity better than $\pm$ 10 K, this means between the hottest and coldest point inside the chamber a maximum difference of 20 K is allowed. The installation of many thermocouples in such a small furnace chamber is too difficult, that's why the ceramic rings are used. Fig. 13 shows the temperature distribution for the ranges 1300 °C to 1700 °C and 10 bar to 75 bar.
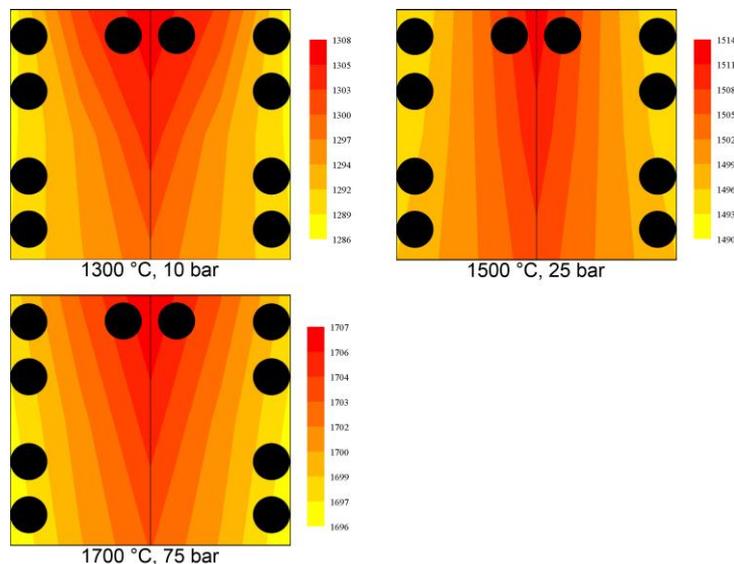
Fig 13 shows the temperature distribution for temperatures between 1300 °C and 1700 °C within the pressure range from 10 bar to 75 bar.

The cross sections of the heating elements are shown in black colour. It can be clearly recognized that the temperature has a maximum at the backside because of the additional heater. When pressure and temperature are increased the accuracy is improved, at 1700 °C and 75 bar the maximum difference is 11 K, while at 1300 °C and 10 bar 22 K difference are observed. This can be explained by improved thermal conductivity at higher pressures and temperatures (fig. 4). Finally an accuracy of approx. $\pm$ 10 K could be confirmed for this furnace, provided that operation under nitrogen or air is done. Measurements under argon atmosphere were not done, however a higher difference is expected because of the lower thermal conductivity of argon (0.017 W / m · K) in comparison to air or nitrogen (0.026 W / m

· K, both values at room temperature).
For each atmosphere the temperature uniformity could be optimized by an additional heater at the front side, however charging would become more difficult.

Conclusion

The general aim of these experiments was the investigation of the relations between the effective thermal conductivity of porous insulation materials and different practical factors like temperature, gas atmosphere and pressure. General guidelines, how to install the correct heating power depending on the mentioned parameters, were formulated using a high pressure furnace. This makes it easier to design similar furnace types, because until now there did not exist reliable results. Under- and overdimensioning heating power were often the consequence.

Furthermore the fact can be recorded that the temperature accuracy of ± 10 K is feasible, better values require optimization at the heating elements' arrangement or at the basic furnace design, e. g. top or bottom loader.

These measurements generated useful knowledge for the design of a 60 bar high pressure furnace for 2000 °C maximum temperature under nitrogen or argon atmosphere (fig. 14). A better temperature accuracy (lower than ± 10 K) was the result of the design as top loader with heating elements installed along the cylinder perimeter. In order to reduce heat up time (app. 45 minutes) 60 kW heating power were installed for 5 liters useful volume in combination with graphite fibre insulation. This corresponds to 12 kW/liter, the above mentioned theory calculates 9,5 kW/liter. Although the graphite fibre insulation is highly effective, a water cooling power of 17 kW is required, as was shown in experiments.

Fig. 14 Rubistar Special in toploader version. On the left side the water cooling unit

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