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Thermal conductivity of non-isotropic materials measured by various methods

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Abstract. High-temperature thermal conductivity of insulating material is usually measured by application of the steady-state calorimeter method or the transient hot-wire method. However, when applied to non-isotropic materials, the methods yield results which show systematic differences depending on orientation of the material during the measurement. In this contribution results are presented for one and the same material measured between room temperature and 1300 °C in all three instruments (according to the steady-state plate, the steady-state cylinder, and the transient hot-wire methods). Additional experiments are carried out in these instruments with various fibre orientations. Differences between the measured conductivities are found to exceed 100% in the high-temperature range. The results are discussed and supplemented by raster electron microscope (REM) and differential thermal analysis (DTA) of the fibre material and numerical studies of the temperature fields inside the facilities.

1 Introduction

Thermal conductivity is an important criterion for the proper choice of insulation materials. The solid components of the porous insulation give the material more or less its mechanical stability, whereas the material choice is dependent on the applicable temperature and the desired form, which in turn is dependent on the structure of the fibres or filling material. The opened or closed cavities in the insulation material are gas filled, typically with air, and with a decrease in pressure an increase of the ability to insulate can be expected.

The effective thermal conductivity is strongly linked to the molecular thermal conductivities of gas and solids, the ratio of their volume (porosity), and geometric alignment and the present radiation. To be able to analyse and separate these parameters a series of physical/mathematical models have been developed, which are partly extremely complicated and are based on a wide range of material characteristics. A calculation from such data alone is not possible without a measurement of the effective thermal conductivity.

It is known that different measurement methods on the same material deliver a wide variety of results. This makes the comparison of the such results extremely difficult and leads to a degree of uncertainty in the thermal conductivities presented by insulation manufacturers in the form of tables, graphs, and comparison formulae.

2 Measurement methods

Over the past 200 years a large number of measurement methods have been developed, which vary widely in their complexity, to measure the thermal conductivity with an appropriate degree of precision. All these methods are based on solutions of Fourier's differential equation for heat conduction along with such boundary conditions as are easy to achieve, yielding standardised (DIN, ASTM, ISO, etc) steady-state and transient measurement methods with specific time and cost factors.

2.1 *Transient hot-wire method*

The most widely used nonsteady method is the transient hot-wire method. A thin wire is embedded in the test material which is at constant temperature before the experiment starts. The test is conducted by heating the wire electrically. Heat is released in the radial direction (ie two-dimensionally in Cartesian space) into the surroundings and the wire temperature climbs with a transient which is more or less steep depending on the thermal conductivity of the test medium. This method is standardised according to DIN EN 993-14, ASTM C 1113-90, and ISO/DIS 8894-2 (DIN 1998; ASTM 1990; ISO/DIS 1990), and it is applied in three forms: single wire, parallel wires, and crossed wires setups.

This method should only be applied to dense materials [in the VDI-Richtlinie 2055 (1994) a density greater than 500 kg m^{-3} is given] up to maximum temperatures between 1250 and 1600 °C, but not to poorly conducting nonceramics (with $\lambda < 0.5$ W m^{-1} K^{-1}), and application to fibrous materials is said to be a problem (see the ASTM standard): "in general it is difficult to make accurate measurements on anisotropic materials, particularly those containing fibers, and the use of this test method for such materials should be agreed between the parties concerned".

Hot-wire measurements have the advantage that they can be conducted with relatively low material costs and within a short period of time. A suggested requirement of this method is that the thermal conductivity of the tested materials is isotropic, ie not directionally dependent. Because the measured thermal conductivity is a result of a two-dimensional average heat flow the application of this value is of limited benefit for practical purposes. To save measurement time one often does not wait until there is an even temperature throughout the sample. In such a case the test material is constantly heated up or cooled down in a furnace with short measurement phases. This has proven to be reliable if there are no heat sources or heat sinks due to reactions in the course of the temperature rise, a condition which is not fulfilled in many cases.

2.2 *Steady-state methods*

Steady-state methods are typically very time and cost consuming. Two opposite surfaces of the sample material (in the form of a plate or a hollow cylinder) are fixed at different temperatures. One must wait until a stationary temperature field has developed uniformly throughout the sample, a procedure which can take many hours to achieve. A particularly difficult problem is the correct determination of the heat flow density through the sample and the exact temperature difference.

The plate method is standardised in DIN 52 612 (DIN 1979) in a relative simple version, with a maximum temperature between 200 and 1000 °C depending on the complexity of the setup. The US standard for plate measurements at high temperatures is based on ASTM C 201 (ASTM 1993b) (for refractories) with special conditions given for insulating fire bricks (in ASTM C 182; ASTM 1993a), refractory bricks (ASTM C 202; ASTM 1993c), and unfired monolithic refractories (ASTM C 417; ASTM 1993d).

2.3 *Comparison of methods in literature*

A glance at the published literature shows that the measured thermal conductivity of high-temperature thermal insulation materials is dependent on which method was used to measure it. This fact was first stated by Koltermann (1961) and explained with the principal uncertainties of the measurements. For the preparation of the standard for the hot-wire method for ceramic materials during the beginning of the 1970s, the possible sources of errors were investigated by a round-robin test with seven research laboratories using fire bricks (classes 23 and 28, temperatures up to about 1000 °C) and the results were presented, for example by Eschner et al (1974). It was reported that the hot-wire method measured higher thermal conductivities than the plate method (typically by about 10% to 15%). Davis and Downs (1980) and also Hagemann and Peters (1982) concluded that the results of the hot-wire method were due to a non-isotropy in the material and were a sort of 'average' between the plate method results for a sample measured in two directions. Davis and Downs questioned the applicability of the hot-wire method for such materials: "some modification of the method for use with anisotropic materials seems necessary particularly for insulating fibres". Dietrichs (1987) reported that vacuum-formed fibrous materials tested with the hot-wire method had up to 20% higher values than the same material tested with the plate method. There is also a large difference in results for calcium silicate materials from the two methods (up to 33%) (Schlegel 1988).

One can conclude that all measurements with the hot-wire method result in thermal conductivities 10% to 30% larger than those measured with the plate method and the application of the hot-wire method on non-isotropic materials is very problematic.

3 Our measurements

The lack of comparable measured results on fibrous materials, especially at high temperatures, was the motivation for a research project on materials which have a range of definite directional dependence of the thermal conductivity.

3.1 *Material and measurement protocol*

Tables 1 and 2 give an overview of the examined materials and the applied measurement methods. Calcium silicate which has a directionally independent thermal conductivity has been used for comparison and three non-isotropic ceramic fibre mats of different bulk densities were chosen for these measurements. Heating up the fibrous mats above 1000 °C resulted in an exothermic reaction and the build-up of pseudospinel. When the fibres were heated above 1200 °C, the amorphic ground materials underwent a crystalline modification to mullite depending on the heating duration and actual temperature. This process results in a shrinkage of the materials and an increase of density. Figure 1 shows a microscopic view (REM) of the ceramic fibres (a) before and (b) after an exposure

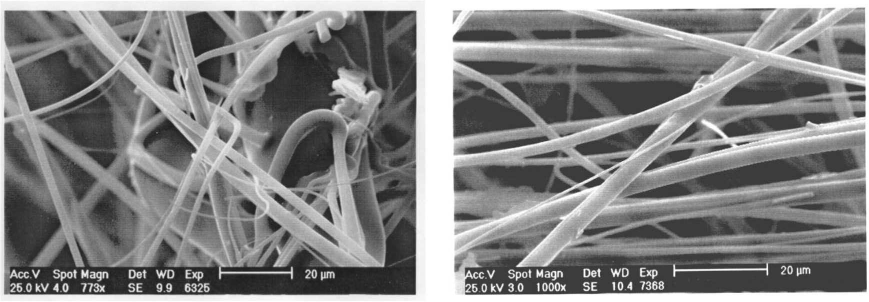
Table 1. Examined materials.

	Ceramic fibre mat	Calcium silicate
Composition	52% Al_2O_3, 48% SiO_2	41% CaO, 42% SiO_2
Classification temperature	1400 °C	900 °C
Bulk density	104/116/158 kg m^{-3}	220 kg m^{-3}
Porosity	96.3%/95.8%/94.3%	92.9%
Pore size:		
average		0.25 μm
range		0.2 to 0.3 μm
Fibres		
diameter	2 to 5 μm	
length	40 to 80 mm	
Structure		
DTA	exothermic reaction at 1006 °C	none
x-ray phase analysis	amorphous ⇒ mullite	xonotlithe ⇒ wollastonite

Table 2. Applied measurement methods (ASTM 1993b; DIN 1998).

	Method	Reference	Temperature/°C	Atmosphere	Dimensions/mm
1	Plate	Bock	20–80	air	250 × 250 × 70 (max.)
2	Plate	ASTM C 201	300–1400	air	300 × 300 × 120 (max.)
3	Cylinder		400–1450	argon	12/60 (r_i/r_a) × 180
4	Hot wire*	DIN EN 993-14	20–1450	air	2 × (250 × 125 × 65)

*The hot-wire measurements were conducted at the Institut für Silikattechnik (Institute for Silicate Technology) at the Bergakademie Freiberg.



(a) (b)

Figure 1. Microscopic view of (REM) of the ceramic fibre mats (a) before and (b) after exposure to a maximum temperature of 1300 °C.

of the material to the maximum temperature of about 1300 °C. It is hard to see any difference between the fibre structures. A comparison of measurements conducted with increasing and decreasing temperatures brought almost identical thermal conductivities for the respective temperatures.

3.2 *Results for calcium silicate*

Figure 2 shows the accumulated measurement results for the calcium silicate material (bulk density 220 kg m^{-3}). The typical increase of the effective thermal conductivity with temperature is noticeable. This is due to the strong effects of radiation at the higher temperatures. The results of the measurements conducted with the plate, hot-wire, and cylinder methods can all be correlated with one trend line within a spread of ±10% which is almost within the measurement tolerances, which have been analysed and calculated to be ±3%, ±4%, and ±5% for steady-state cylinder and plate instruments and the transient hot-wire instrument, respectively.

It is interesting to note that the values supplied by the manufacturer correspond with measured results at the lower temperatures (up to 300 or 400 °C), but they are well below the measured results for the higher temperature. The manufacturer's values are said to be measured with the hot-wire method and then given for plate geometry (by what means has not been outlined). This result shows clearly a certain criticism of values provided in the advertisements of insulation manufacturers. The measurements will be continued concentrating on a possible discovery of a miniscule non-isotropy in this material.

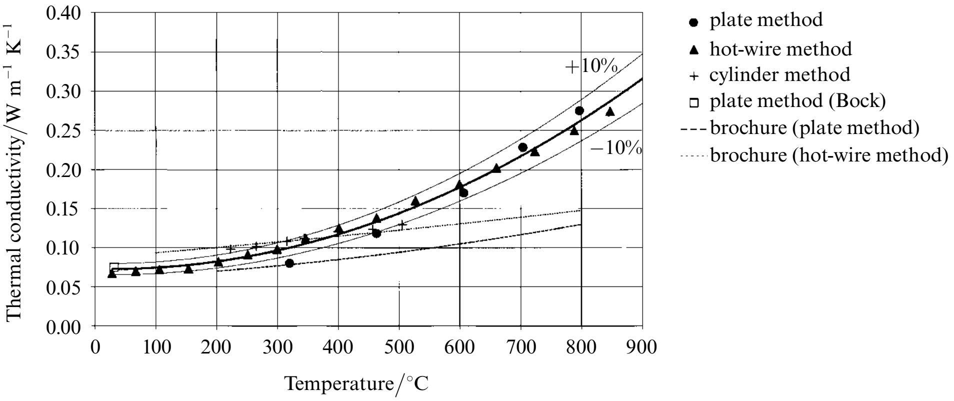


Figure 2. Thermal conductivity results for calcium silicate.

3.3 *Results for ceramic fibre mat*

It is extremely important when handling fibrous mats, especially when loading into a test apparatus, that one does not inadvertently change the bulk densities. Besides this, insulation delivered from the manufacturers sometimes has large deviations in the local density of test pieces cut out from a large mat.

3.3.1 *Measurements with the plate method.* Figure 3 shows a schematic of the plate measurement apparatus used for the measurements. It is the third generation of apparatus built by the Freiberg University of Technology (Bergakademie) since the middle of the 1970s. A 300 mm × 300 mm sample is placed at the lower boundary of an electrically heated furnace. The measurement zone is the 100 mm × 100 mm core of the sample. Underneath the sample is a calorimeter for the exact measurement of the heat flow which is encased in a system of several layers of shield calorimeters. It is especially important that in the measurement core the streamlines of heat flow are exactly parallel. To ensure this the development of the measurement apparatus was accompanied by a finite-element

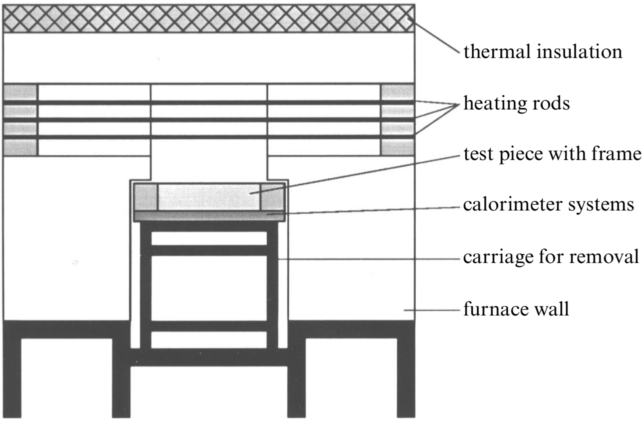


Figure 3. Schematic drawing of the plate test facility.

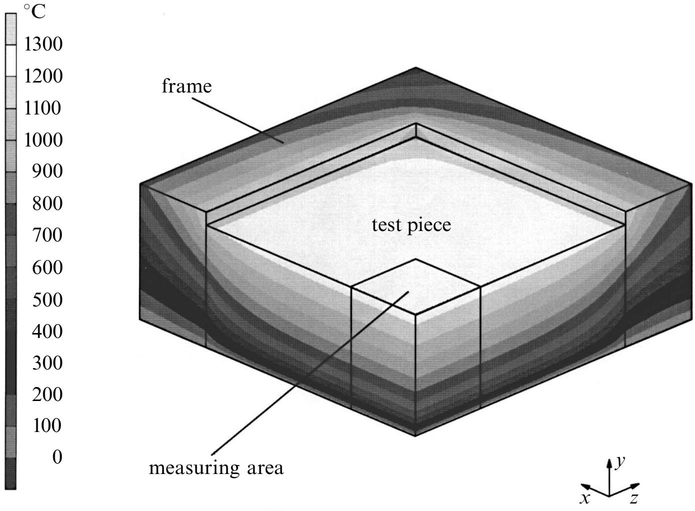


Figure 4. Temperature field in the test section of the plate facility (finite-element calculation at furnace temperature 1300 °C).

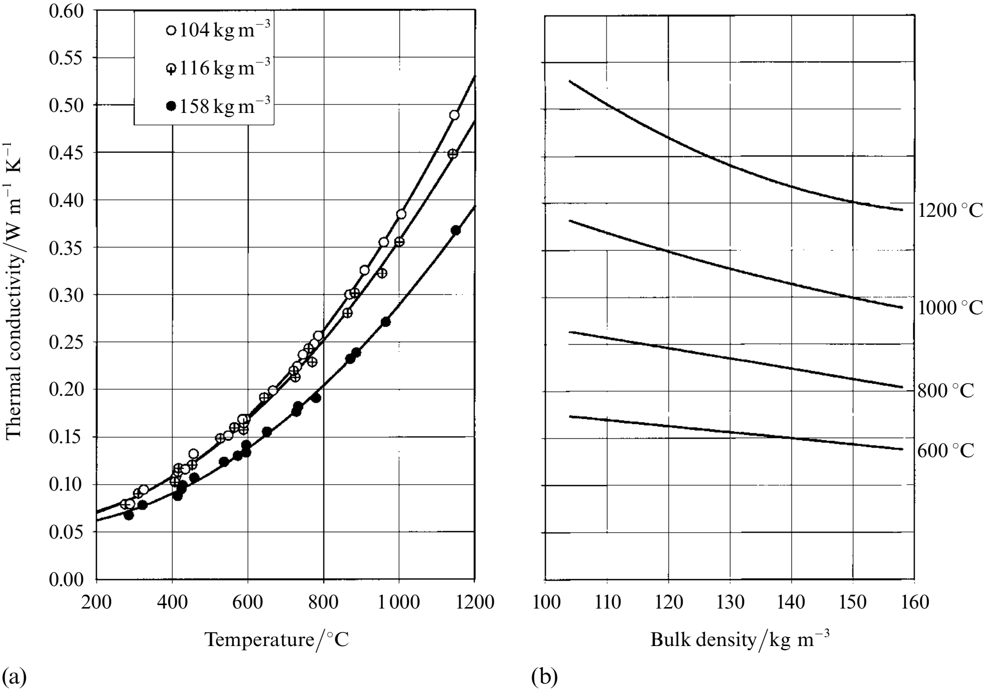


Figure 5. Thermal conductivity results for ceramic fibre mats of various bulk densities (measured by the plate method).

simulation which provided insight into the temperature distribution through the sample and its surroundings (figure 4). The temperature is measured in three levels each with five thermocouples which have been calibrated at regularly designated time intervals.

The measurements are conducted when steady-state conditions are achieved. This means test durations of several hours for one measurement. Typically a test series is made up of about 20 measurements taken with increasing then decreasing temperature taking a total time of up to two weeks. Figure 5a shows three such test series for ceramic fibre mats of different densities. One can clearly see that in this radiation-dominated region the thermal conductivity is lower for higher densities. In figure 5b this phenomenon is clearer as a result of the interpolation values for the chosen temperatures and this shows how important it is to know the raw density, especially at the higher temperatures.

3.3.2 *Expectations of other methods.* The measurements up to this point have been conducted on the insulation materials in their normal installation direction. In the y-direction (figure 6) perpendicular to the main fibre orientation the measured thermal

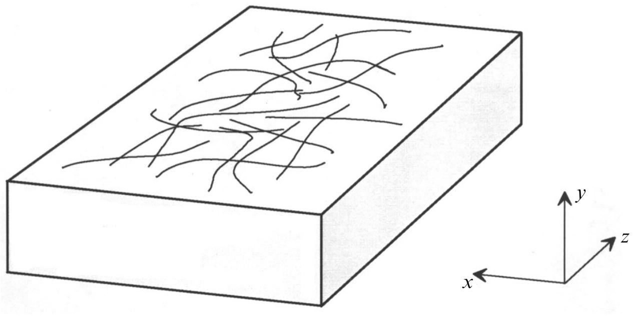
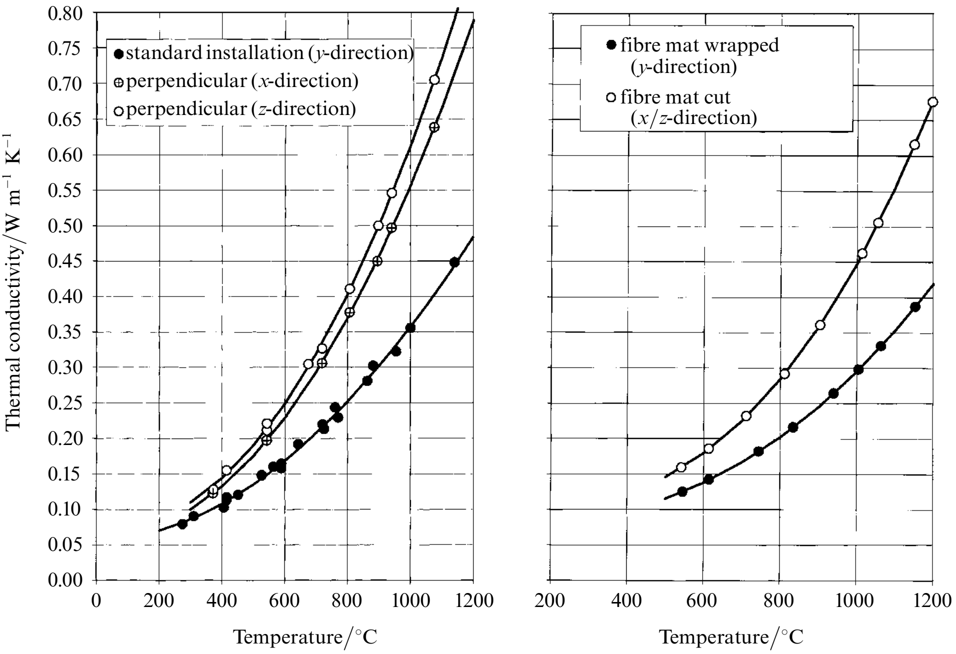


Figure 6. Definition of x, y, z directions.

Table 3. Overview of the relevant heat-flow directions.

No.	Heat-flow direction	Qualitative thermal conductivity	Expected order
Plate method			
1.1 Mats	y		4
in 'normal' position	λ_y	low	
1.2 sliced, layered, and set on end	x		1
	λ_x	much higher	
1.3 similar but in the other direction	z		1
	λ_z	much higher	
Cylinder method			
2.1 Mats	x/z		1
cut out	$\lambda_x \ldots \lambda_z$	rather high	
2.2 wrapped (carefully)	y		4
	λ_y	low (like 1.1)	
Hot-wire method			
3.1 Hot-wire between 2 mats	x/y		3
in main direction	$\lambda_x \ldots \lambda_y$	rather low	
3.2 Hot-wire between 2 mats	y/z		2
across the main direction	$\lambda_z \ldots \lambda_y$	higher	
3.3 Hot-wire perpendicular to the mats	x/z		1
	$\lambda_x \ldots \lambda_z$	rather high (like 2.1)	



(a) (b)

Figure 7. Thermal conductivity results for ceramic fibre mats: orientation effect as measured by (a) the plate method and (b) the cylinder method.

conductivity is lower than in the x and z directions. Due to manufacturing conditions, there is a somewhat noticeable dominant direction in either the x or z direction. The comparison between the expected results of these directions is shown in table 3 (with $\lambda_z \approx \lambda_x \gg \lambda_y$). From this table it is clear that in non-isotropic material (naturally not only fibrous mats) differences in the thermal conductivity must be present.

Figure 7a shows the plate-measured thermal conductivity for the three directions for the ceramic fibre mat with a bulk density of $\rho = 116$ kg m^{-3}. In the 'normal' installation position (in the y-direction) the heat flows perpendicular to the fibre length and the measured conductivities are lower in this direction compared to the other ones.

3.3.3 *Measurements with the cylinder method.* With the cylinder method, a hollow cylinder sample is electrically heated from the inside. The outside surface is cooled with the help of a calorimeter system. Thermocouples are set at two different radii. Hollow cylinder samples are simple to manufacture if the test material is solid. For fibre mats, however, the manufacture of these samples is quite difficult and must be done with great care. To measure the thermal conductivity perpendicular to the fibre length (in the y-direction) the sample is wrapped around the inner core. To measure the thermal conductivity parallel to the fibre length, samples have to be cut out of the material to form rings. The rings are then stacked around the inner core to fill the hollow space in the apparatus. For the analysis of these measurements three points have to be considered: (i) the sample has to be placed in an argon atmosphere which makes it necessary to have a conversion formula for air; (ii) by wrapping the material around the core there is an increase in density (up to 123 kg m^{-3}) which makes it necessary to have a conversion formula to the original density (116 kg m^{-3}), and (iii) it is difficult to correlate the results to a given temperature because of the nonlinear temperature relationship and the large temperature difference. It is therefore necessary to apply the process of Bolte (1957) which allows the thermal conductivity to be calculated as a function of the temperature. The results of the cylinder method are shown in figure 7b. One can see that the cut-out sample has a much higher thermal conductivity than the wrapped one.

3.3.4 *Measurements with the hot-wire method.* The 'normal' application of the hot-wire method for fibrous materials involves placing the crossed wires in either the x or z direction between two mats of sample material. In both cases the result is an 'average' value consisting of components from all directions in which the y-direction components could possibly have little influence. A third possible method is to thread the wire through a stack of mats. The results should be a higher measured thermal conductivity. Figure 8a shows, however, that the results for all directions are more-or-less the same. Therefore, the fibre direction does not have a measurable influence on the thermal conductivity of this low-density material. The radiation transport, which dominates the total heat transfer process, is highly affected by the heat flow density close to the wire (which is usually much higher than in the case of plate instruments). This has been found from additional experiments with various electric currents. The results have to be extrapolated to zero heat flow density, a procedure which is typically avoided in practice.

The results also show that it was not possible to achieve a stable measurement at temperatures around 1000 °C where the expected exothermic reaction takes place. Interpretable results were first possible at temperatures above 1150 °C.

3.3.5 *Comparison of methods.* The comparison in figure 8b shows that the plate method in the y-direction, ie the normal installation direction, has an approximately 20% higher measured thermal conductivity compared to the cylinder method measurements with wrapped-around fibre mats while this is more-or-less in line with the values provided by the manufacturer. The variations of the transient hot-wire method compare up to

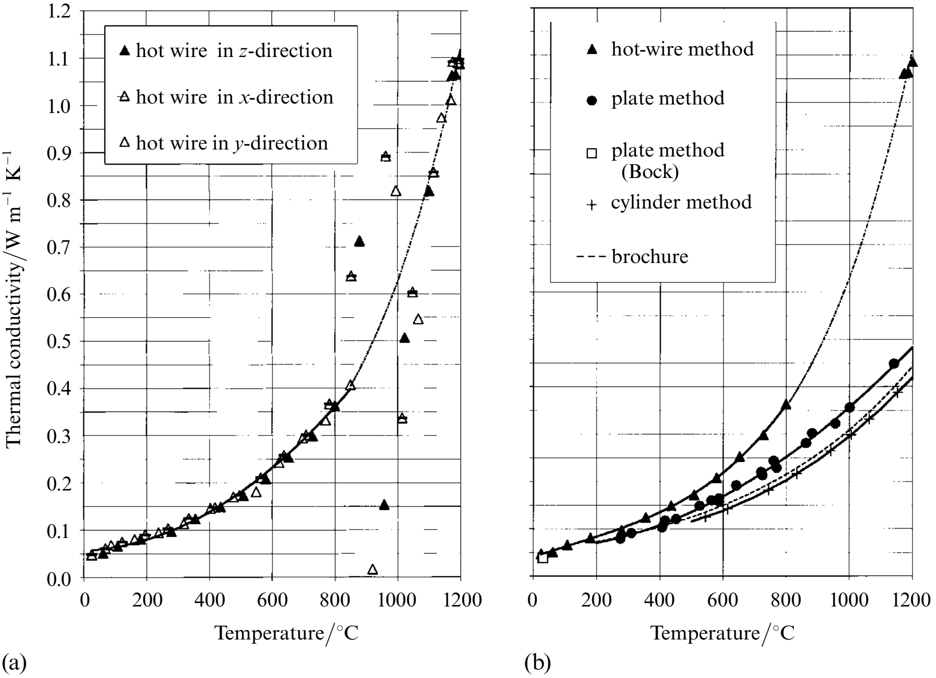


Figure 8. Thermal conductivity results for ceramic fibre mats: (a) orientation effect as measured by the hot-wire method, and (b) comparison of the various methods.

around 400 °C with the two stationary methods. At higher temperatures, however, the hot-wire results begin to exceed the conductivities from the other methods. This drastic increase can only be explained through the strong influence of the heat radiation. Compared to the plate measurements, at 800 °C the hot-wire method resulted in 50% larger values and at 1200 °C, 120% larger values.

4 Summary and conclusions

The thermal conductivities of the isotropic calcium silicate material and the ceramic fibre mats which are strongly non-isotropic were measured by different methods. All three methods provided uniform results for the calcium silicate material. This was, however, not the case for the ceramic fibre mats. The measured thermal conductivities with the plate and cylinder methods were largely dependent on the fibre direction of the sample, which was not the case for the hot-wire method. Conductivities measured with the hot-wire method can exceed the plate and cylinder method measurements by more than 100% for the "normal" installation direction. The hot-wire results at higher temperatures with their extraordinarily large measured values are difficult to explain other than through the influence of heat radiation. This phenomenon provides a challenging theme for further research.

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References

ASTM, 1990, ASTM C 1113-90, "Standard test method for thermal conductivity of refractories by hot wire" (West Conshohocken, NJ: American Society for Testing and Materials)

ASTM, 1993a, ASTM C 182, "Standard test method for thermal conductivity of insulating fire brick" (West Conshohocken, NJ: American Society for Testing and Materials)

ASTM, 1993b, ASTM C 201, "Standard test method for thermal conductivity of refractories" (West Conshohocken, NJ: American Society for Testing and Materials)

ASTM, 1993c, ASTM C 202, "Standard test method for thermal conductivity of refractory brick" (West Conshohocken, NJ: American Society for Testing and Materials)

ASTM, 1993d, ASTM C 417, "Standard test method for thermal conductivity of unfired monolithic refractories" (West Conshohocken, NJ: American Society for Testing and Materials)

Bolte W, 1957 *Brennstoff-Wärme-Kraft* **9** 373 – 375

Davis W R, Downs A, 1980 *Trans. J. Br. Ceram. Soc.* **79** 44 – 52

Dietrichs P, 1987 *Silikattechnik* **38** 349 – 353

DIN, 1979, DIN 52612, "Wärmeschutztechnische Prüfungen—Bestimmung der Wärmeleitfähigkeit mit dem Plattengerät" (Berlin: Deutsches Institut für Normung; Beuth Verlag)

DIN, 1998, DIN EN 993-14, "Prüfverfahren für dichte geformte feuerfeste Erzeugnisse—Bestimmung der Wärmeleitfähigkeit nach dem Heissdraht- (Kreuz-) Verfahren" (Berlin: Deutsches Institut für Normung; Beuth Verlag)

Eschner A, Grosskopf B, Jeschke P, 1974 *Tonind. Ztg.* **98** 212 – 219

Hagemann L, Peters E, 1982 *Interceram* **31** 131 – 135

ISO/DIS, 1990, ISO/DIS 8894-2, "Refractory materials—determination of thermal conductivity—hot wire method" (Geneva: International Organisation for Standardisation)

Koltermann M, 1961 *Tonind. Ztg.* **85** 399 – 407

Schlegel E, 1988, pesonal communication

VDI-Richtlinie 2055, 1994 *Wärme- und Kälteschutz für betriebs- und haustechnische Anlagen* (Düsseldorf: VDI-Verlag)