32nd International Thermal Conductivity Conference
20th International Thermal Expansion Symposium
April 27–May 1, 2014
Purdue University, West Lafayette, Indiana, USA

A New Facility for the Experimental Investigation on Nano Heat Transfer between Gas Molecules and Ceramic Surfaces

D. Bayer, doreen.bayer@iwtt.tu-freiberg.de, U. Gross, ulrich.gross@iwtt.tu-freiberg.de, and R. Wulf, rhena.wulf@iwtt.tu-freiberg.de
Department of Thermodynamics and Heat Transfer, Technical University Bergakademie Freiberg
K. Raed, k.raed@roeber-institut.de
Röber Institut Petkus GmbH

ABSTRACT

Since the last decade, the interest has risen in nanoscaled technological products, which have advantages through their size effect. The size effect also plays a significant role in the area of micro- and nanoscale heat transfers. Many applications were developed using this effect, such as nanostructured porous media, e.g. Aerogels or ceramics. This experimental work is focused on the determination of thermal accommodation coefficients (TAC) on ceramic surfaces considering several influencing factors. TAC is influenced by temperature, kind of gas, kind of wall material, roughness, and contamination with adsorbed gas layers. To determine TAC, a parallel plate apparatus is used, although most of the measurements in the past have been arranged with the hot wire method. Using the parallel plates apparatus is advantageous due to its simpler machining of specimens and to analyze the effect of roughness. The apparatus is build similar to the well-known guarded hot-plate method using an unidirectional heat flux through the gas layer due to a small temperature gradient, which is provided by two different heat foils. The measurements take place between 30 and 100°C, 10^{-4}, and 1 mbar.

Keywords: thermal accommodation coefficient, thermal conductivity, ceramics, nano heat transfer.

1. INTRODUCTION

Since the last decade an interest has risen in nanoscaled technological products that have advantages through their size effect. This size effect also plays a significant role in the area of micro- and nanoscale heat transfer. Many applications were developed using this effect such as nanostructured porous media, e.g., Aerogels. Stark and Fricke (1993) and many others have shown that the effective thermal conductivity of highly porous ceramic materials decreases significantly with a decreasing pressure. Especially in porous ceramics with micro- and nanopore sizes or fibrous insulations, this behavior is found. The dependency of the effective thermal conductivity on pressure or size effect is called Smoluchowski effect (see Raed and Gross, 2007) or, respectively, Knudsen effect (see Raed and Gross, 2007) or noncontinuum effect (see Rader et al., 2004). Several models were developed to predict the gas thermal conductivity as a contribution to the effective thermal conductivity in dependency of gas pressure or Kn-number, which is the ratio of the free mean path and a characteristic length scale (Dulnev, Muratova, Tribel, Madzhidov, & Safarov, 1986; Kaganer, 1969; Schlünder and Tsotsas, 1988). Considering these models, the influence of a certain coefficient (the thermal accommodation coefficient α) on the resulting gas thermal conductivity λ_{gas} is significant. This has been specified by Kaganer (1969), Wawryk and Rafalowicz (1988), and Wakao and Vortmeyer (1971), respectively, as (see also Figure 1):

$$\lambda_{\text{gas}} = \frac{\lambda_0}{1 + 2\beta Kn} \tag{1}$$

$$\beta = \frac{2k}{\kappa + 1}\frac{2-\alpha}{\alpha} \tag{2}$$

$$\beta = \frac{19}{6}\frac{2-\alpha}{\alpha} \tag{3}$$

and

$$\beta = 2\frac{2-\alpha}{\alpha} \tag{4}$$

where λ_{gas} is the gas thermal conductivity at lower pressure, λ_0 the gas thermal conductivity at 1 bar, Kn is the Kn-number, β a variable, k a constant, and κ the ratio of specific heats. Equations (1)–(4) have been evalutated for $\alpha = 0.1$ and 0.8 and the results are shown in Figure 1. An extraordinary effect is obtained for $Kn = 0.1$, for example, regarding the ratio of $\lambda_{\text{gas}}/\lambda_0$ which jumps from about 0.2 to 0.7. This significant rise shows the importance of the thermal accommodation coefficient for the effective thermal conductivity of highly porous materials.

DOI: 10.5703/1288284315549

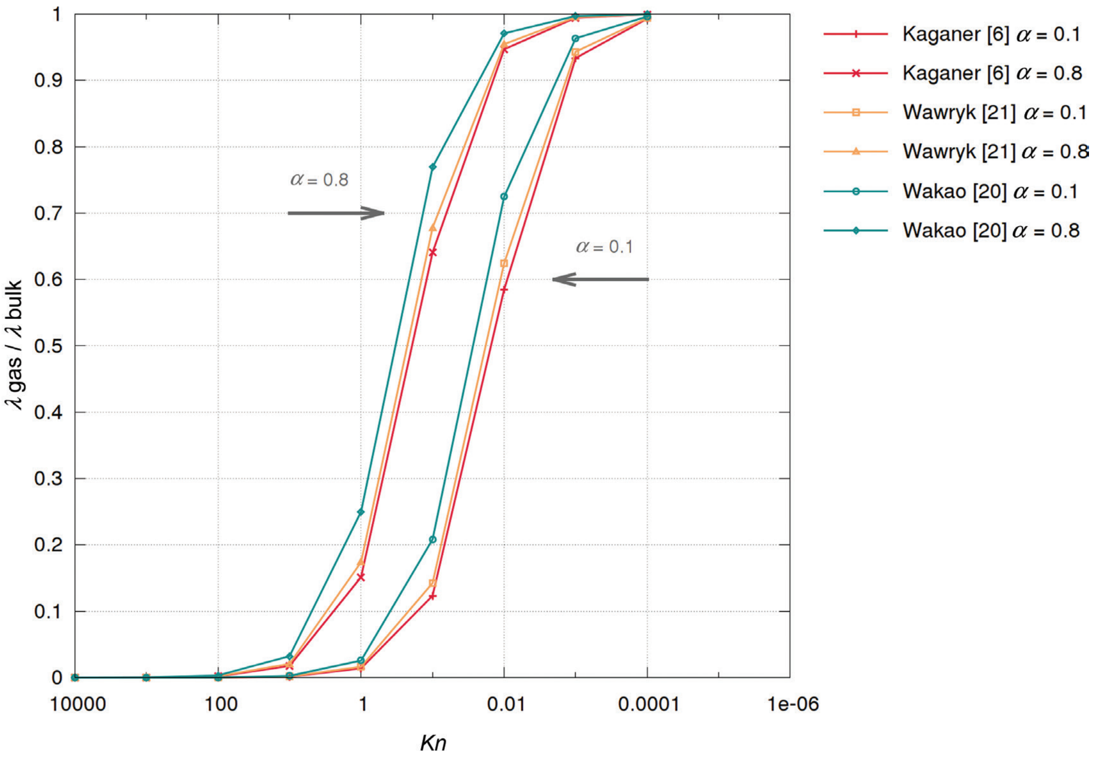

Figure 1. Dependency of gas thermal conductivity over Kn-number on two different α using the models of Kaganer (1969), Wawryk and Rafalowicz (1988), and Wakao and Vortmeyer (1971).

The thermal accommodation coefficient α was introduced by Knudsen (1911) and it describes the efficiency of heat transfer during collision between a molecule and a wall. Therefore, it is the relation of the amount of energy that is actually transferred and that which could be transferred, if the molecule and the wall would have come to a thermal equilibrium (Saxena and Joshi, 1989):

$$\alpha = \frac{\Delta E_{\text{real}}}{\Delta E_{\text{ideal}}} \tag{5}$$

The thermal accommodation coefficient is influenced by several factors, such as temperature, kind of gas, kind of wall material, its roughness and its contamination with adsorbed gas layers. Especially, the contamination of adsorbed gas layers has shown a significant influence on α (Rolf, 1944). In the literature, only a few experimental results are available for ceramic materials, as the measurements of thermal accommodation coefficients were focused on metallic surfaces, which are used in space or nanotechnology applications. This work aims to measure the thermal accommodation coefficients for ceramic materials and to analyze the different influencing factors.

2. THEORY

The thermal accommodation coefficient becomes important when the gas pressure or the pore size is significantly reduced and the Kn-number exceeds a limiting value of about $Kn = 0.01$. Therefore, the Kn-number is used as an indicator for the state of the gas. A commonly used classification has been suggested by Schaaf and Chambre (1958) as follows[1]:

- $Kn > 10$ free molecular motion;
- $10 > Kn > 0.1$ transition regime;
- $0.1 > Kn > 0.01$ temperature jump regime;
- $0.01 > Kn$ continuum.

Figure 2 shows, however, that the lower limit of the continuum exhibits a great dependency on the thermal accommodation coefficient. Therefore, it should be considered to limit the continuum state to $Kn = 0.001$ (α-limit). For predicting the heat transfer in dependency of the thermal accommodation coefficient, different models were developed because of the changing state of gas. In the following, the models of free molecular motion, transition regime, and continuum are described considering the heat transfer between two parallel plates, as an example.

2.1 Continuum

In the continuum state the gas thermal conductivity is nearly independent of gas pressure showing only a slight increase with increasing pressure. Heat is

---

[1] In this work, the transition and temperature jump regimes are combined to the transition regime.

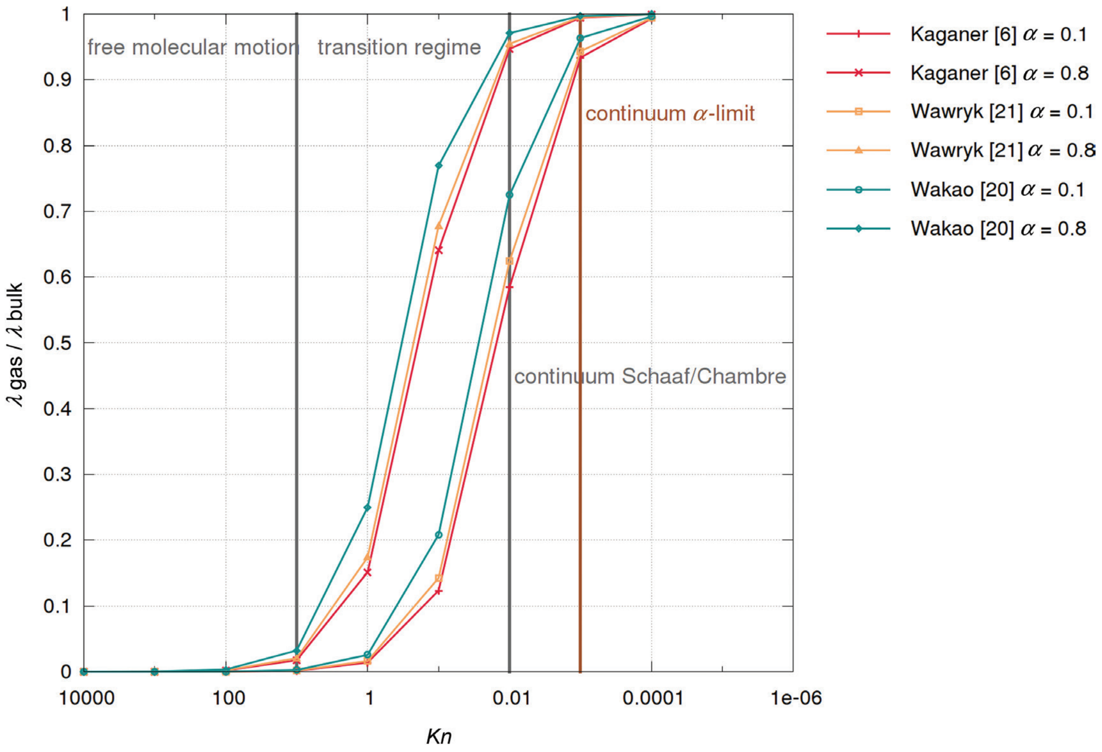

Figure 2. Gas states in dependency of Kn-number using the model of Kaganer (1969), calculated for helium at 25°C.

transfered because of intermolecular collisions that play the major role. It follows the well-known Fourier Law as

$$\dot{q} = -\lambda \frac{dT}{dx} \tag{6}$$

yielding to

$$\dot{q}_{\mathrm{C}} = -\lambda_0 \frac{T_{\mathrm{hot}} - T_{\mathrm{cold}}}{d} \tag{7}$$

for a system of parallel plates, where q_{C} is the heat flux in the continuum state, λ_0 the gas thermal conductivity in the continuum state, d the thickness of gas layer, i.e., specimen distance, and $T_{\mathrm{hot}} - T_{\mathrm{cold}}$ the temperature difference across the gas layer.

2.2 Free molecular motion

In this state the gas is strongly rarefied and characterized by $Kn > 10$. Intermolecular collisions are significantly reduced and the collisions between gas molecules and the wall are mainly responsible for the heat transfer with a strongly increased role of the thermal accommodation coefficient. According to Springer (1971), the transfered heat can be described by the following equation which is derived from the definition of the thermal accommodation coefficient [Equation (5)]:

$$\dot{q}_{\mathrm{FM}} = \frac{\alpha_1\alpha_2}{\alpha_1+\alpha_2-\alpha_1\alpha_2}\, p\left(T_{\mathrm{hot}} - T_{\mathrm{cold}}\right) \frac{C_v - R/2}{\sqrt{2\pi M R T_{\mathrm{gas}}}} \tag{8}$$

where $\dot{q}_{\mathrm{FM}}$ is the heat flux in the free molecular regime, α_1 and α_2 the thermal accommodation coefficients of the plates, p is the gas pressure, $T_{\mathrm{hot}} - T_{\mathrm{cold}}$ the temperature difference along the gas layer, C_{v} the specific heat at constant volume, R the specific gas constant, M the molar mass and T_{gas} the gas temperature.

2.3 Transition regime

In the transition regime, the heat transport process is governed by intermolecular and gas molecule to wall collisions. This situation is much more difficult to predict requiring a solution of the Boltzmann equation (Rader et al., 2004). Especially for parallel plates, several solutions have been published because of the simple geometry of parallel plates (Springer, 1971). The most promising solution was found by Lees and Liu (1962) in comparison with experimental data. Following Springer (1971), it further agrees best with the Monte–Carlo solution. By using Direct Simulation Monte Carlo method, Bird (1994) showed more recently that the solution of Lees and Liu agrees very well with the numerical results. However, the solution of Lees and Liu (1962) is only valid under the assumptions of a small temperature gradient and a monatomic gas. Springer (1971) extended the solution of Lees and Liu (1962) to the following Equation (9) for arbitrary thermal accommodation coefficients:

$$\frac{\dot{q}_{\mathrm{TR}}}{\dot{q}_{\mathrm{FM}}} = \frac{1}{1+\dfrac{4}{15}\dfrac{1}{Kn}\left[\dfrac{\alpha_1\alpha_2}{\alpha_1+\alpha_2-\alpha_1\alpha_2}\right]} \tag{9}$$

According to Shermans interpolation (Shermans, 1963)

$$\frac{1}{\dot{q}_{TR}} = \frac{1}{\dot{q}_{C}} + \frac{1}{\dot{q}_{FM}} \tag{10}$$

It is also possible to rewrite Springers (1971) solution [Equation (9)] in the following form:

$$\frac{\dot{q}_{TR}}{\dot{q}_{C}} = \frac{1}{1+\frac{15}{4}Kn\left[\frac{\alpha_1+\alpha_2-\alpha_1\alpha_2}{\alpha_1\alpha_2}\right]} \tag{11}$$

with $\dot{q}_{TR}$, $\dot{q}_{FM}$, and $\dot{q}_{C}$ as the heat fluxes in the transition, the free molecular, and the continuum regimes, respectively.

3. EXPERIMENTAL SETUP

At first, it is necessary to determine in which regimes (transition and/or free molecular) the measurements with parallel plates can be carried out. Therefore, the upper and lower limits have been determined (see Figure 3). The upper limit is given by surface adsorption of gases that increases α and leads to wrong measurement values. The lower limit is defined by the lowest heat flux that can be measured satisfactorily through the measurement system (1mW). It can be seen in Figure 3, that most of the measurements will be carried out in the transition regime ($10 > Kn > 0.01$), but it is also possible to measure in the free molecular regime ($Kn > 10$).

3.1 Requirements

An evacuable device is required for the measurement of thermal accommodation coefficients which becomes important at $Kn > 0.001$ and, in addition, different gas atmospheres (Helium, Argon, Nitrogen) will be considered. Therefore, several essential requirements have to be fulfilled by the measurement device:

1. *Constant low pressure and defined gas atmospheres*: A test chamber is needed, which can be evacuated down to high vacuum conditions allowing an exchange of the gas atmosphere. As shown by Rolf (1944), adsorbed gas molecules have a significant influence on the thermal accommodation coefficient measurements leading to increased results. Therefore, it is also important to keep the pressure in the vacuum range, where surfaces are only covered with adsorbed gas molecules by 1% of the whole area.
2. *Minimized volume of the vacuum chamber*: Evacuation of the vacuum chamber is a time consuming procedure because of its volume and the desorption process of physically and chemically adsorbed gas molecules. The former adsorption type can be overcome by pressure reduction, but for the chemically adsorbed gas molecules, a higher activation energy

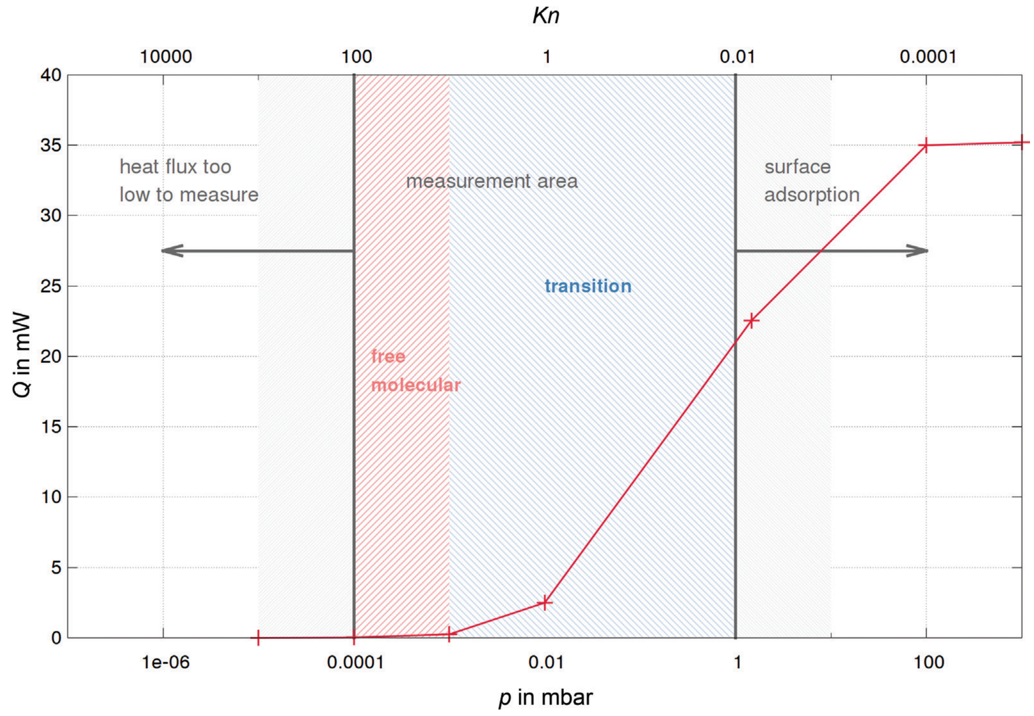

Figure 3. Upper and lower limit of measurement area.

has to be provided by heating (bake out). This extends the whole evacuation process even more. Furthermore, the procedures of pressure reduction and bake-out have to be repeated several times to make sure that surfaces are clean and the outgassing rate of dissolved molecules in solid materials is reduced to a minimum level.

3. *Avoiding porous media as insulation materials*: Because of the heterogeneous structure of porous materials, their high specific surface with increased capillar activity, and their tortuosity, the outgassing rate is much higher. The evacuation and desorption times are excessively extended.
4. *Short duration of specimen exchange*: The modification and exchange of a specimen (e.g., placement of thermocouples) shall not take longer than 1 day to keep the usability of the measurement device on a good level.
5. *Operation temperatures up to 200°C*: The measurements are performed from almost room temperature up to 200°C to examine temperature influences on the thermal accommodation coefficient. No insulation and cooling mechanisms will be installed inside the vacuum chamber for keeping the desorption time short.

3.2 Basic concept

The concept of the accommodation coefficient measurement (ACM) device is based on the principle of a unidirectional heat flux through a plain gas layer that is enclosed between two parallel plates (see Figure 4). This kind of arrangements has been applied earlier in similar experimentals (Peck and Lokay, 1958; Rader et al., 2004; Teagan and Springer, 1967). The method is advantageous, as theproducibility of ceramic specimens and the installation in the ACM device are easier than the mostly used hot-wire method. Highly porous ceramics are very brittle and they are hardly measurable and installable with small geometrical dimensions as it is needed for most other measurement methods. As it is metrologically not possible to measure heat flow rates or even thermal accommodation coefficients in micro- or nanopores inside of ceramic materials, the pores will be exposed to the gas layer. Therefore, the measurement of α on the surface of the ceramic material is considered to be the same as it is within the pores. Of course, it has to be considered that, at reduced pressures, not only gas conduction takes place. Radiation is omnipresent and has to be determined separately. As it was done by Rader et al. (2004), the radiative heat transfer will be determined at the lowest pressure to neglect gas conduction. Later, the radiative heat flux will be subtracted from the measured heat flux at higher pressures.

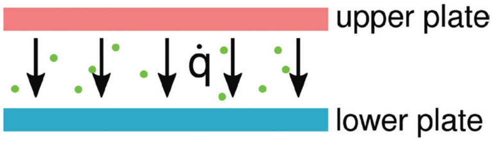

Figure 4. Unidirectional heat flux between parallel plates.

3.3 Assembly of the measuring device

The ACM device was planned and build at the Technische Universität Bergakademie Freiberg, Saxony, Germany. It consists of an upper and lower assembly with heat foils and specimens. It is positioned within a vacuum chamber to reach low enough pressures where the influence of α is recognizable. The scheme of ACM device is given in Figure 5. In the following, several parts of the ACM device will be explained further as they fulfill some important tasks in the assembly.

For the upper plate assembly extensive considerations have been done to realize the measurement principle of unidirectional heat flux. Therefore, a system of guarding heat foils was developed to create an isothermal area around the metering heat foil forcing the heat flux through the gas layer toward the ceramic specimen (see Figure 5). All heating devices were manufactured as heat foils made of polyimide. This is advantageous because of the small dimensions of the circular heaters leading to an improved temperature distribution along the heat foil. Furthermore, heat foils are extremely thin, which makes lateral losses negligible and does not expand the inner assembly unnecessarily. All heat foils are embedded between metal plates (e.g., reference specimen) for keeping them at the desired position.

The metering heat foil consists of a central metering area (A) which is surrounded by a guarding area (B) for minimization of lateral heat losses (see Figure 6). Both of them are controlled separately. The metering heat foil has an overall diameter of 100 mm. The central path delivers the heat flow for the accommodation coefficient measurements and it is intended to keep its temperature as uniform as possible. To determine the diameter of the metering area (A), numerical simulations were carried out in dependence on the heat losses of the ceramic specimens which have a certain height, where lateral losses can occur. As a result, using a specimen height of 20 mm agrees with the limit of a maximum temperature deviation along the metering area of 10 mK within a circle with a diameter of 30 mm. Choosing lower specimen heights is more difficult because of the temperature measurement which will be explained in what follows. Higher specimen heights are less advantageous as lateral heat losses increase.

Guarding heat foils are needed to create an isothermal area in the upper assembly above the gas layer. In

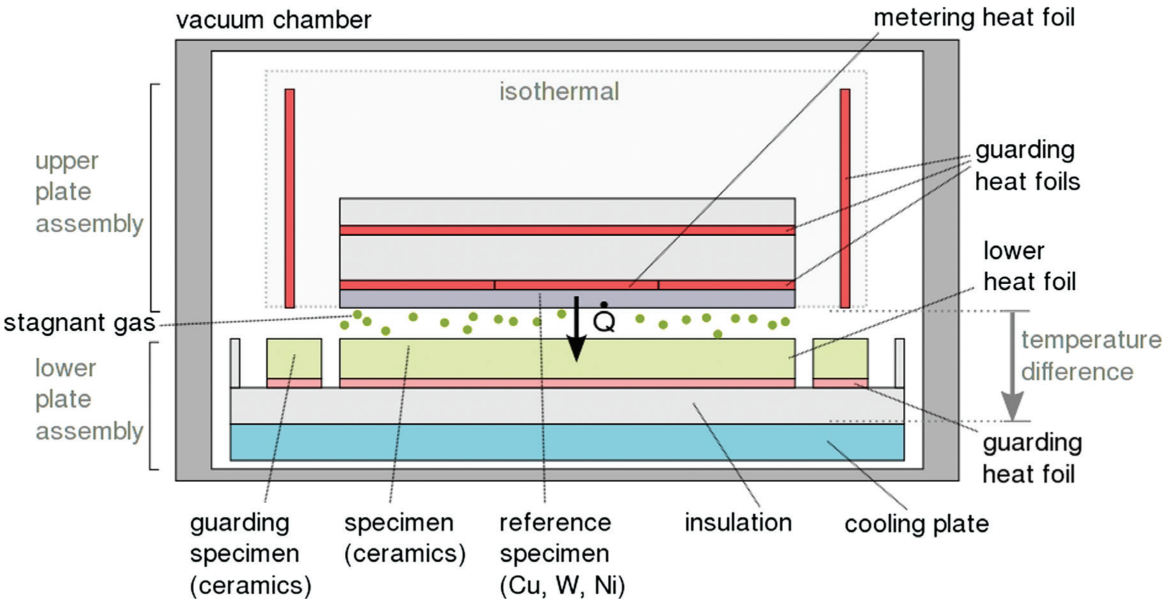

Figure 5. Scheme of concept of upper and lower assemblies.

Figure 5, it is shown that the guarding heat foils are positioned radial and axial around the metering area. They are controlled to a zero temperature difference to the metering heat foil.

In the lower plate assembly, the guarding heat foil system is following the existing temperature difference along the ceramic specimen. Therefore, a ring-shaped heater is placed concentric to the lower heat foil which is supposed to produce the same temperature difference through the guarding specimen. The latter one consists of the same material as the ceramic specimen. Owing to this, the temperature difference between the specimen and the guarding specimen should be significantly smaller than without the guarding specimen reducing lateral heat losses.

For the temperature measurements, thermocouples type K with a diameter of 1 mm are used and produced at once to eliminate systematic errors. To position the thermocouples within the ceramic specimen, the thinnest possible bore-hole to the center of the specimen can be made with a 1.1-mm drilling head. Therefore, a diameter of 1 mm for all used thermocouples was chosen. As the surface temperature of the ceramic specimen is needed to calculate α in the transition and free molecular regime, four thermocouples are placed within the ceramic specimen. They measure the linear temperature difference at four points which is later used to extrapolate the surface temperature. Placing a thermocouple directly on the surface would falsify the surface quality leading to wrong measurement results of α.

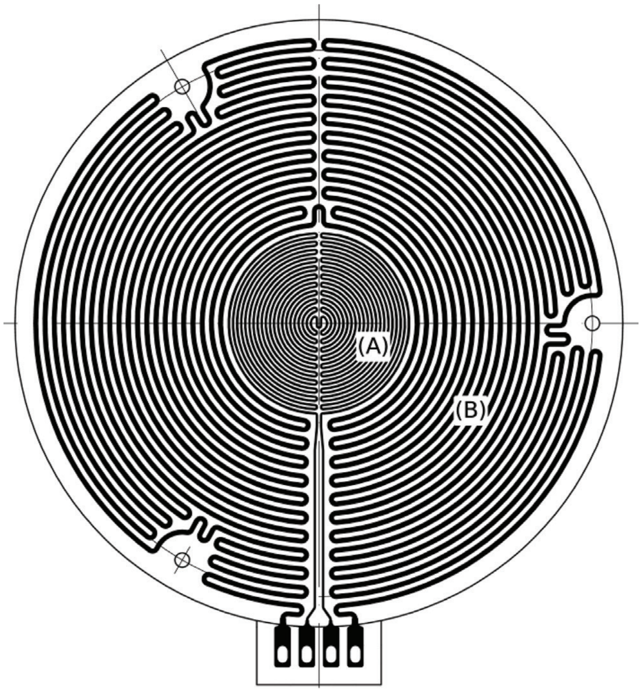

Figure 6. Metering heat foil with metering area (A) and guarding area (B).

For insulation, a dense ceramic material was chosen with a solid thermal conductivity that is relatively low in comparison to the materials of all other components. It is called steatite and is mainly based on magnesium silicate with a thermal conductivity of 2–3 W/mK (Verband der Keramischen Industrie e.V, 2014). Steatite is placed between metering heat foil and upper guarding heat foil and between the lower heat foil and

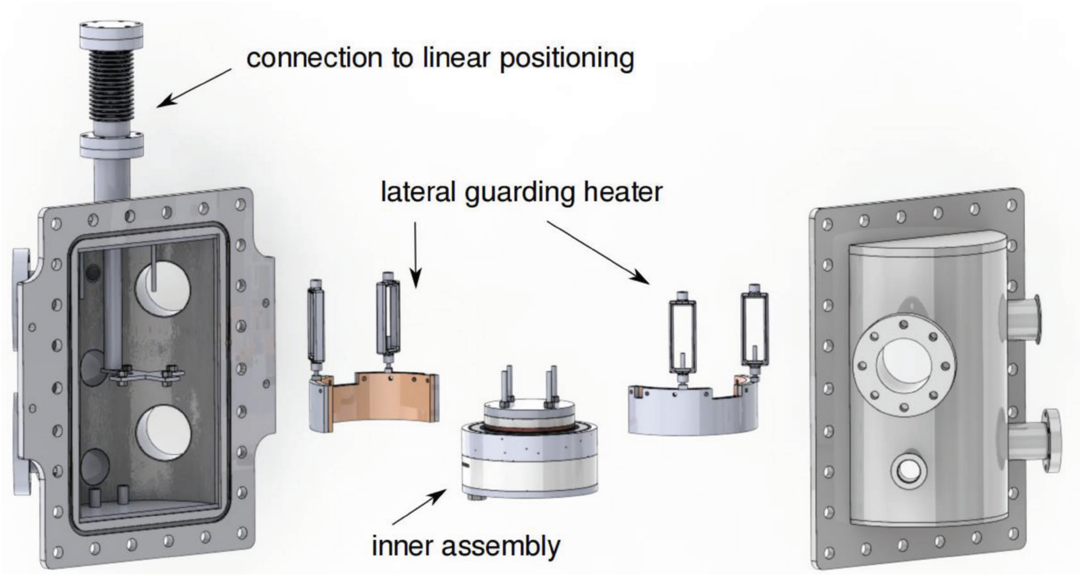

Figure 7. Exploded view of ACM.

cooling plate as well. In the latter place, it additionally supports the positions of the specimen and the guarding specimen to keep them in concentric positions.

A cooling plate with inside water flow is used to provide a heat sink in the thermal concept of the ACM device. In prior experimental works such as Bienkowski (1962) and Teagan and Springer (1967), a single spiral was used as cooling plate. This design was used as the basis for further enhancement of the cooling plate via numerical simulations. Owing to this a double-spiral was developed that offers an appropriate radial temperature distribution and does not have an influence on the radial temperature distribution along the specimen facing the gas layer. The cooling plate has a diameter of 150 mm.

In Figure 7, an exploded view of the ACM device is given showing the vacuum chamber, the connection to the linear positioning of the inner assembly, and lateral guarding heaters. The lateral heaters are held separately to avoid any thermal connection to the inner assembly.

4. SUMMARY

In this article it is shown, that the thermal accommodation coefficient has an important impact on the thermal conductivity of gases. An examplary calculation by using the models of Kaganer (1969), Wawryk and Rafalowicz (1988) and Wakao and Vortmeyer (1971) showed a significant deviation of the thermal conductivity with variation of α confirming the importance. Furthermore, the measurement concept of the ACM device for determination of the thermal accommodation coefficient on ceramic materials is explained. The method of parallel plates – similar to the guarded hot plate method – is used, which exhibits some advantages like the examination of the influence of the wall roughness and simpler machining of specimens. Careful consideration had to be done to create a guarding system of heaters that forces the heat flux through the gas layer unidirectional. In addition, it is possible to examine the influences of different temperatures (up to 200°C), temperature differences, and gases.

REFERENCES

Bienkowski, G. K. (1962). *Kinetic theory analysis of problems in the transition range of Knudsen numbers* (PhD thesis). Massachusetts Institute of Technology, Cambridge, MA.

Bird, G. A. (1994). *Molecular gas dynamics and the direct simulation of gas flows*. Oxford: Oxford University Press.

Dulnev, G. N., Muratova, B. L., Tribel, T. V., Madzhidov, K. H., & Safarov, M. M. (1986). Method of calculating the thermal conductivity of porous granular materials with metal filler in different media. *Journal of Engineering Physics, 51*, 940–943.

Kaganer, M. G. (1969). *Thermal insulation in cryogenic engineering*. Jerusalem: Israel Program for Scientific Translations.

Knudsen, M. (1911). Die molekulare wärmeleitung der gase und der akkommodationskoeffizient. *Annalen Der Physik, 4*(34), 593–656.

Lees, L., & Liu, C. Y. (1962). Kinetic-theory description of conductive heat transfer from a fine wire. *The Physics of Fluids, 5*, 1137–1148.

Peck, R. E., & Lokay, J. E. (1958). Accommodation coefficients of hydrogen, helium, air and argon for chrome surfaces at reduced pressures. *Journal of Chemical and Engineering Data, 3*, 37–42.

Raed, K., & Gross, U. (2007). Review on gas thermal conductivity in porous materials and Knudsen effect. In *Proceedings of the 29th International Thermal Conductivity Conference*, Birmingham, Alabama, USA, June 24–27, 2007.

Rader, D., Trott, W., Torczynski, J., Gallis, M., Castanada, J., & Grasser, T. (2004). *Microscale rarfied gas dynamics and surface interactions for euvl and mems applications*. Technical Report. Sandia National Laboratories, Albuquerque, New Mexico, USA.

Rolf, P. (1944). The accommodation coefficient of helium on platinum. *Physical Review, 65*(5), 185–189.

Saxena, S. C., & Joshi, R. K. (1989). *Thermal accommodation and adsorption coefficients of gases*. New York, NY: Hemisphere Publishing Corporation.

Schaaf, S. A., & Chambre, P. L. (1958). Flow of rarefied gases. *Fundamental of Gasdynamics, 3*, 687–739.

Schlünder, E. U., & Tsotsas, E. (1988). *Wärmeübertragung in Festbetten, durchmischten Schüttgütern und Wirbelschichten*. New York, NY: Georg Thieme Verlag Stuttgart.

Sherman, F. S. (1963). *Rarefied gas dynamics*. New York, NY: Academic Press.

Springer, G. S. (1971). Heat transfer in rarefied gases. *Advances in Heat Transfer, 7*, 163–218.

Stark, C., & Fricke, J. (1993). Improved heat-transfer models for fibrous insulations. *International Journal of Heat Mass Transfer, 36*(3), 617–625.

Teagan, W. P., & Springer, G. S. (1967). Plane layer type apparatus for gas thermal conductivity measurements. *The Review of Scientific Instruments, 38*(3), 335–339.

Verband der Keramischen Industrie e.V. (2014). *Magnesiumsilikate (c 200)*, Technical report Selb, Germany, 3.

Wakao, N., & Vortmeyer, D. (1971). Pressure dependency of effective thermal conductivity of packed beds. *Chemical Engineering Science, 26*, 1753–1765.

Wawryk, R., & Rafalowicz, J. (1988). The influence of residual gas pressure on the thermal conductivity of microsphere insulations. *International Journal of Thermophysics, 9*, 611–625.