Newly Designed Apparatus for Measuring the Angular Dependent Surface Emittance in a Wide Wavelength Range and at Elevated Temperatures up to 1400°C

M Rydzek, T Stark, M Arduini-Schuster and J Manara
Bavarian Center for Applied Energy Research (ZAE Bayern),
Am Hubland, 97074 Wuerzburg, Germany
E-mail: rydzek@zae.uni-wuerzburg.de

An optimized apparatus for measuring the angular dependent surface emittance up to elevated temperatures has been designed. This emittance measurement apparatus (EMMA) is coupled to a Bruker Vertex 70v FTIR-spectrometer, so that a wavelength range from about 2 µm up to 25 µm is accessible. The central part of the new apparatus is a double walled, stainless steel vessel which can be evacuated or filled with various gases or with air. Inside the vessel a cylindrical tube furnace is pivot-mounted on a system of discs, for automatically rotating up to an angle of 180°. This allows both, the measurement at different detection angles (0° to 85°) and a consecutive measurement of sample and black-body reference without ventilating and opening the pot. The aim of this work is to present the newly designed emittance measurement apparatus which enables the determination of the angular dependent spectral emittance of opaque samples at temperatures up to 1400°C. Next to the setup of the apparatus, the measurement results of various materials are presented at different detection angles.

1. Introduction

Heat transfer, especially at the surface boundaries mainly consist on three significant mechanisms, such as heat conduction, convection and thermal radiation. The thermal radiative transport in particular is characterised by the spectral reflectance, transmittance as well as absorbance or respectively emittance of the investigated material.

The emittance is given by the ratio of the radiation intensity emitted by a surface and the radiation intensity emitted by a black-body at the same temperature and wavelength. The exact knowledge of the spectral surface emittance is vitally important for many physical and technical applications, especially when the radiative heat transport needs to be specified or when temperature measurements have to be performed contactless.

At ambient temperature, the contribution of radiation to the total heat transport lies in the same order of magnitude than the heat transfer caused by natural convection in air. With increasing temperatures, the contribution of thermal radiation strongly dominates the total heat transport. Due to that fact, the radiative material properties are decisive in high temperature applications.

1 To whom any correspondence should be addressed.
Hence, for the development of new functionalized high-emitting materials and components, the determination of the surface emittance, especially at elevated temperatures, is necessary.

In general, the surface emittance of a certain material can be achieved by different methods, which can be subdivided into calorimetric and radiometric techniques. In case of the radiometric measurement techniques, the radiative properties of the sample are determined, whereupon a differentiation between indirect and direct radiometric techniques needs to be done.

Using an indirect method, the conservation of energy is used to calculate the spectral emittance from the measured reflectance and transmittance values. In contrast, by applying a direct radiometric measuring technique, as used in this work, it is possible to directly detect the thermal radiation emitted by a sample surface that is heated to a certain temperature. Therefore, a comprehensive study of the radiative properties of a certain material in a wide temperature range can be performed.

2. Theoretical Background

2.1. Emittance

The spectral directional emittance $\varepsilon_{d,\lambda}$ is defined as the ratio of the spectral directional intensity emitted by a surface $i_{d,em}$ at a given temperature $T$, wavelength $\lambda$ and emission angle $\theta$ and the spectral directional intensity emitted by a black-body $i_{d,bb}$ at the same temperature $T$ and the same wavelength $\lambda$ [1]:

$$\varepsilon_{d,\lambda}(\theta, T) = \frac{i_{d,em}(\theta, T)}{i_{d,bb}(T)}.$$

(1)

The spectral directional intensity emitted by a black-body is independent of the emission angle. Additionally it can be assumed, that the spectral directional emittance only depends on the polar angle $\theta$ and is invariant due to changes of the azimuth angle $\phi$. Based on the energy conservation for opaque samples follows:

$$1 = \alpha_{d,\lambda}(\theta, T) + \rho_{dh,\lambda}(\theta, T),$$

(2)

with $\alpha_{d,\lambda}$ being the spectral directional absorbance and $\rho_{dh,\lambda}$ the spectral directional-hemispherical reflectance. By using Kirchhoff’s law:

$$\alpha_{d,\lambda}(\theta, T) = \varepsilon_{d,\lambda}(\theta, T),$$

(3)

together with the equation for the conservation of energy one can derive the well-known coherency for opaque samples:

$$\varepsilon_{d,\lambda}(\theta, T) = 1 - \rho_{dh,\lambda}(\theta, T).$$

(4)

Moreover, the directional total emittance $\varepsilon_{d,total}$ gives the total amount of thermal radiation at a given temperature, emitted by a surface. It can be calculated by integrating the spectral directional emittance $\varepsilon_{d,\lambda}$ over all wavelengths with the Planck-function of the black-body $i_{d,bb}$ as a weight function:

$$\varepsilon_{d,total}(\theta, T) = \int_{\lambda_1}^{\lambda_2} \varepsilon_{d,\lambda}(\theta, T) \cdot i_{d,bb}(T) \cdot d\lambda.$$

(5)

Typical values for the directional emittance of electrically conducting, as well as non-conducting materials as a function of the emission angle can be obtained by theoretical calculations using the Maxwell equations with appropriate boundary conditions like shown in [1] for example.
2.2. Direct Radiometric Measurement Method

Within this work, the emittance is determined by a direct radiometric method [2] which detects the thermal radiation emitted by a sample surface that is heated to a certain temperature. For measuring the spectral directional emittance $\varepsilon_{d,\lambda}$ of a surface with the surface temperature $T_S$ and the emission angle $\theta$, not only the radiation emitted by the surface itself has to be taken into account. Moreover, the radiation impinging from the surrounding and being reflected by the sample surface into the view of sight has to be considered [3].

Therefore, the measured spectral directional intensity $i_{\lambda,\text{meas}}$ at a surface temperature $T_S$ and a temperature of the black-body surrounding $T_U$ is given by the sum of the intensity emitted by the sample and the intensity coming from the black-body surrounding and being partly reflected by the sample surface into the view of sight:

$$i_{\lambda,\text{meas}}(\theta, T_S, T_U) = \varepsilon_{d,\lambda}(\theta, T_S) \cdot i_{\lambda,\text{bb}}(T_S) + \rho_{\text{bd},\lambda}(\theta, T_S) \cdot i_{\lambda,\text{bb}}(T_U).$$

Hence, the existence of a black surrounding with a constant and heterogeneous temperature distribution is fundamental for measuring the emittance with a high accuracy by using a direct radiometric method.

Considering the law of reciprocity:

$$\rho_{\text{bd},\lambda}(\theta, T) = \rho_{\text{db},\lambda}(\theta, T),$$

together with the energy conservation for opaque samples (equation (2)) and Kirchhoff’s law (equation (3)), the spectral directional emittance of the sample surface can be obtained as follows:

$$\varepsilon_{d,\lambda}(\theta, T_S) = \frac{i_{\lambda,\text{meas}}(\theta, T_S, T_U) - i_{\lambda,\text{bb}}(T_S)}{i_{\lambda,\text{bb}}(T_S) - i_{\lambda,\text{bb}}(T_U)}.$$  

(8)

Here, $i_{\lambda,\text{meas}}$ is the measured intensity as defined in equation (6) and $i_{\lambda,\text{bb}}$ is the intensity emitted by a black-body at the temperatures $T_S$ and $T_U$, respectively.

Thus, the spectral directional emittance of the sample surface can be derived from an intensity measurement together with two associated theoretically calculated black-body spectra. Due to additional influences of the spectrometer and the measurement setup, the detected intensity $i_{\lambda,\text{detected}}$ is not identical with the measured intensity $i_{\lambda,\text{meas}}$. The measured intensity $i_{\lambda,\text{meas}}$ gives the intensity from the sample surface as defined above, whereby the detected intensity $i_{\lambda,\text{detected}}$ gives the value displayed by the apparatus. This value depends not only on the sample and its surrounding but also on the spectrometer and its configuration. To obtain the measured intensity $i_{\lambda,\text{meas}}$ which is needed for further evaluations, a correction term is necessary:

$$i_{\lambda,\text{detected}} = C_1(\lambda) \cdot i_{\lambda,\text{meas}} + C_2(\lambda).$$  

(9)

In fact, the measured intensity $i_{\lambda,\text{meas}}$ can be calculated from the detected intensity $i_{\lambda,\detected}$ by use of two calibration functions named $C_1(\lambda)$ and $C_2(\lambda)$. Thus, before measuring the sample, the spectrometer has to be calibrated [4].

By assuming the temperature of the spectrometer to be constant, both calibration functions $C_1(\lambda)$ and $C_2(\lambda)$ are only depending on the wavelength. Within this assumption, $C_1(\lambda)$ considers the influence of the different components of the spectrometer and the measurement setup (such as lenses, mirrors, detector, beam-splitter, electronic parts, etc.) on the detected intensity. The calibration function $C_2(\lambda)$ represents the detected background radiation of the spectrometer and the measurement setup itself [5].
The two calibration functions \( C_1(\lambda) \) and \( C_2(\lambda) \) can be obtained by determining the intensity of a black-body source \( i_{\lambda, \text{bb}, \text{detected}} \) with a well-known theoretically calculated intensity \( i_{\lambda, \text{bb}} \) at two different temperatures. Additionally, the calibration performance can be optimized by measuring three black-body spectra at three different temperatures [4] and also by statistically averaging the values to minimize the uncertainty of the experiment [6]. In this case one obtains:

\[
C_1(\lambda) = \sum_{\kappa \leq 1}^{3} a_{k, l} \frac{i_{\lambda, \text{bb}}(T_i) - i_{\lambda, \text{bb}, \text{detected}}(T_i)}{i_{\lambda, \text{bb}, \text{detected}}(T_k)} ,
\]

and

\[
C_2(\lambda) = \sum_{m=1}^{3} b_m \left[ \frac{i_{\lambda, \text{bb}}(T_m)}{C_1(\lambda)} - i_{\lambda, \text{bb}, \text{detected}}(T_m) \right] .
\]

The uncertainty of the calibration is given as the difference between the theoretically derived intensity of the black-body and the corrected intensity:

\[
i_{\lambda, \text{bb}, \text{corrected}} = i_{\lambda, \text{bb}} - C_2(\lambda) .
\]

Therefore, the relative uncertainty of the calibration can be calculated from:

\[
\sigma_{i_{\lambda, \text{bb}}} = \frac{i_{\lambda, \text{bb}, \text{corrected}} - i_{\lambda, \text{bb}, \text{corrected}}}{i_{\lambda, \text{bb}}} .
\]

Within this work, the relative uncertainty of the calibration of the used emittance measurement apparatus (EMMA) is well below 1% and thus has only little influence on the total uncertainty of the performed measurements.

3. Experimental setup

3.1. Emittance measurement apparatus (EMMA)

The self-designed emittance measurement apparatus used within this work consists of a double-walled stainless steel vessel with a diameter of 500 mm and a height of 300 mm which can be evacuated or filled with different gases. The application of different gaseous atmospheres (such as argon or other inert gases) or working in vacuum helps to eliminate the absorbing effects of water (H\(_2\)O) and carbon dioxide (CO\(_2\)) which are present in air and additionally limits the oxidation effect on the sample surfaces at higher temperatures.

The temperatures of the top, bottom and side walls of the vessel can be adjusted by a thermostat and therefore homogeneously tempered to a constant value. Therefore, a cooling liquid is pumped through the double-walled vessel, which ensures a stable and uniformly distributed surrounding temperature which is important as the measured intensity is significantly influenced by the temperature of the black-body environment (see equation (6)).

Moreover, the internal walls of the vessel are coated with a highly emitting paint called “Nextel-Velvet-Coating 811-21” from Mankiewicz Gebr. & Co. GmbH, manufactured under license of the Minnesota Mining and Manufacturing (3M) company, which provides a surface emittance of about 0.98 in the infrared spectral region. Due to that, a near to black enclosure with a defined temperature of the surrounding is assured and multiple reflections within the vessel can be neglected.

Inside the vessel a cylindrical tube furnace with a wrapped electric resistance heating wire is pivot-mounted on a system of discs, for automatically rotating up to an angle of 180° (figure 1). This allows both, the measurement at different detection angles (0° to 85° - relative to the surface normal) and a consecutive measurement of sample and black-body reference without ventilating and opening the pot, because the sample and the blackbody are positioned on opposite sides of the tube furnace being simultaneously heated to the desired temperature in the range of 200 °C to 1400 °C. The vessel is
coupled to a Bruker Vertex 70v FTIR-spectrometer using a system of KBr-lenses in order to align the beam focus.

**Figure 1.** Sketch of the emittance measurement apparatus (EMMA) which is coupled to a FTIR-spectrometer (Bruker Vertex 70v) using a system of KBr-lenses. The left picture shows the EMMA where the furnace is positioned for a measurement at a detection angle of 0° (relative to the surface normal). The picture in the middle shows the possibility of angular dependent measurements by rotating the furnace on the system of discs and the picture on the right shows the possibility of consecutively measuring blackbody and sample by rotating the furnace up to an angle of 180°.

Within this setup, metallic or ceramic cylinders with an external diameter of 22-26 mm and a conic cavity serve as black-body reference. The cavity is coated with an exhaust paint which provides an emittance above 0.9. This leads to a total emittance of the cavity which is larger than 0.999 [7]. The impinging intensity originating from the black-body or the sample can be adjusted by two apertures located within the spectrometer. Additionally, the two KBr-lenses are mounted in a separated vacuum chamber which is placed into the optical path between the spectrometer and the vessel.

3.2. Determination of the surface temperatures

The exact knowledge of the relevant temperatures is of significant importance for the determination of the surface emittance and the resulting uncertainty is substantially affected by the accuracies of the measured temperatures.

In this setup, the temperature of the black surrounding is measured by thermocouples fixed on the inner side of the vessel. The temperature of the black-body is detected by two thermocouples placed at different points inside the walls of the cylindrical structure that serves as black-body reference.

For the determination of the surface temperature of the samples, different techniques have been used. In case of the ceramic sample, thermocouples have been fixed on the front and back side of the specimen by use of a sealing strip. In case of the copper sample, two copper-sheets with a thickness of 1 mm have been put together on top of each other, whereupon the thermocouple was fixed in between to achieve a good thermal connection.

For the stainless steel alloy, two holes with a diameter of 1.3 mm and a depth of 15 mm have been drilled in each specimen at a distance of 2 mm and two thermocouples are placed inside these holes. If the sample thickness is significantly smaller than its diameter one can assume that only one dimensional heat transfer occurs. In this case, Fourier’s law can be written as follows:

\[ \dot{q} = -k \cdot \frac{\partial T}{\partial x}. \]  

(14)

Here, \( \dot{q} \) describes the local heat flow and \( k \) stands for the thermal conductivity of the used material. While performing the measurement, the samples are in thermal equilibrium.
4. Results and discussion

Within this work, the emittance of each investigated material was measured at an emission angle of 0° (normal to the surface), as well as at different emission angles up to 85° and the directional total emittance of the samples was calculated according to equation (5). In the angle-range from 0° to 60° an increment of 5° was used. Within the range from 60° to 85° a smaller step size of 2° was chosen, due to a stronger, angle-dependent variation of the emittance in this area.

In the following, the measurement results of a ceramic sample as example for an electrically insulating material, a copper sample as example for an electrically conducting material and an alloy are presented.

4.1. Ceramic sample (TiO$_2$ / Y$_2$O$_3$)

As an example for an electrically insulating material, the measurement results of a ceramic specimen are presented. The sample consists of 60% yttrium oxide (Y$_2$O$_3$) and 40% titanium dioxide (TiO$_2$). For preparing the specimen, the oxides were mixed together and then pressed uniaxial to achieve a green body. After sintering, a wafer-shaped ceramic sample with a thickness of 2 mm and a porosity of 50% has been achieved.

The sample temperature during the measurement was ascertained with a value of 750 °C. Figure 2 shows the spectral emittance in a wavelength range from 6 to 22 microns at this temperature for different emission angles.

To visualize the emittance as a function of the emission angle, the measured values for three different wavelengths (see figure 2) are plotted in a polar diagram (see figure 3). For a wavelength of 7 µm, the spectral emittance is relatively low. At the point of the Christiansen wavelength (12.2 µm) [8], an emittance value of 1 is reached. This value only slightly decreases with increasing wavelengths, so that a value of 0.9 can be detected at 20 microns.
Figure 3 shows the typical behaviour of an electrically insulating material. For detection angles from 0° to about 60°, the spectral emittance shows an almost constant value, whereas for detection angles above 60° a strong slope of the spectral emittance can be detected.

4.2. Copper sample (Cu)
As an example for an electrically conducting material, the measurement results of a copper specimen are presented. Copper exhibits a low specific resistivity of about $1.7 \times 10^{-8} \, \Omega \cdot m$ and a high thermal conductivity of about 400 W/(m·K).

The copper specimen was measured at temperatures of 500 °C and 700 °C in vacuum. Due to the low emittance of copper, the measured intensity is very low. The optimum signal-to-noise ratio was found for a wavelength of 5 µm. Therefore, the examination of the angular dependant emittance was performed only at this wavelength (see figure 4).

Figure 4 shows the typical behaviour of an electrically conducting material. For detection angles from 0° to about 60°, the spectral emittance shows an almost constant and very low value of about 0.03. For detection angles between 60° and 85° a strong increase followed by a rapid decrease of the spectral emittance can be detected. Within this range, a temperature dependant maximum value is visible. At a sample temperature of 500 °C, the maximum value of the emittance was calculated to 0.27. For higher temperatures, the maximum value of the emittance decreases and a value of 0.23 was calculated for a sample temperature of 700 °C.

4.3. Alloy (stainless steel)
At the end, the measurement results of an alloy are presented. Here, a stainless steel composition (V2A-steel) with a fraction of 18 % chromate and 10 % nickel was used as specimen. The resistance to corrosion is achieved by a thin self-passivation layer of the material which protects the steel against a proceeding oxidation.

To make sure that the self-passivation layer is fully formed, the sample was firstly heated in air to 750 °C. Afterwards the measurement was performed in vacuum at the same temperature.

As a result, a spectrally selective behaviour of the emittance was found. For lower wavelengths, the sample approximately behaves like an electrical insulator, whereas for higher wavelengths, the sample shows a metal-like behaviour. The polar diagram exemplarily shows the curve progression for wavelengths of 5 and 14 microns (see figure 5).

Figure 4. Polar diagram of the angular dependant emittance of a copper specimen at a wavelength of 5 microns. The curve progression shows the typical behaviour of a good electrical conductor.
At a wavelength of 14 µm a metal-like emittance with a low value of about 0.1 is measured. An almost linear rising of the emittance can be identified for detection angles between 0° and 80°.
In contrast, the measured emittance at a wavelength of 5 microns shows a constant value of about 0.25 for detection angles between 0° and 50°. Around a detection angle of 80°, both curves show an almost comparable behaviour.

5. Conclusions

In this work, the newly designed apparatus for measuring the angular dependent surface emittance at elevated temperatures has been introduced. With the new EMMA it is possible to immediately measure the spectral directional emittance of a sample surface at different temperatures, angles, as well as under different atmospheres. Measurements on electrically insulating and electrically conducting materials have been shown, which prove the functionality of the measurement apparatus. Additionally, the results indicate the necessity of emissivity measurements at elevated temperatures, as the infrared optical properties significantly depend on temperature.

6. References

[1] Siegel R and Howell J 2010 Thermal radiation heat transfer (Bristol, PA: Taylor & Francis)
[2] Del Campo L, Pérez-Sáez R B, Esquisabel X, Fernández I and Tello M J 2006 Rev. Sci. Instrum. 77 113111-1 – 8
[3] Manara J, Brandt R, Kuhn J, Fricke J, Krell T, Schulz U, Peters M and Kaysser W A 2000 High Temp. – High Press. 32 361-8
[4] Lindermeier E, Haschberger P, Tank V and Dietl H 1992 Appl. Opt. 31 4527-33
[5] Pérez-Sáez R B, Del Campo L and Tello M J 2008 Int. J. Thermophys. 29 1141-55
[6] Wolberg J 2006 Data analysis using the method of least squares (Berlin: Springer)
[7] Brandt R, Bird C and Neuer G 2008 Measurement 41 731-6.
[8] Manara J, Arduini-Schuster M and Keller M H 2011 Infrared Phys. Techn. 395-402

Acknowledgments
This work was carried out with funding by the European Union within the European Metrology Research Programme (EMRP). The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union.