Thermal Management of Heating Processes – Measuring Heat Transfer Properties

F. Raether, J. Baber, H. Friedrich

The design of any heating process is based on a careful thermal management of heat transfer. A necessary prerequisite for this thermal management is the precise knowledge of thermal material properties. Need for improvements exist especially at high temperatures, where measurements of quantities like thermal conductivity or specific heat are difficult. Special thermooptical devices have been developed to provide these data. Concepts and examples are presented in this report. They are compared to customary measuring methods. Regarding greenhouse gas emissions action is required to develop new heating processes for sustainable production of energy intensive products. This includes furnace construction, high temperature components and process parameters. The measurement of heat transfer properties is an important part of this development.

1 Introduction

Heat transfer is the key issue in all thermal processes. Overall, there are three different heat transfer mechanisms: heat radiation, heat conduction and convective flow. Heat radiation is controlled by emissivity of refractories, charge and fluids participating in the high temperature processes. Heat conduction is characterised by thermal conductivity λ or thermal diffusivity a. The first is obtained by simply multiplying thermal diffusivity with the specific heat c_p and density ρ of the respective material:

$$\lambda = \rho \cdot c_p \cdot a \qquad \text{(eq. 1)}$$

Convective flow is taken into account by heat transfer resistances between solids and fluids, which depend on fluid flow. However, the flow itself is not addressed in the current report which concentrates on thermal properties of solid materials.

The design of heating processes involves knowledge of the respective material properties and their dependence on temperature. In the next paragraph general requirements for the measurements are described whereas specific measuring methods are discussed in the next sections.

Generally high accuracy and reproducibility of the measured data is required together with efficient measuring processes. With refractories the coarse microstructure is a problem deteriorating reproducibility of standard measuring methods in many cases. This is illustrated in a simple example where a material consisting of two phases which have a characteristic size S are considered. It is assumed that the two phases are randomly distributed and that the composite properties can be calculated by the rule of mixtures. E.g., the latter applies for specific heat. Then reproducibility can be calculated as a function of measured volume (Fig. 1). In the example the volume fractions of the two phases were both 50 % and the material contrast, i.e. the difference between the specific heat of the two phases was either 10 % or 50 %. It can be seen that for a large characteristic size S or a large material contrast large measuring volumes of 30 cm³ and above are required. On the other hand, very large samples can deteriorate accuracy and efficiency of the measurements, because temperature gradients may occur within the samples during heating up and during measuring. So a compromise is required and sample volumes between 30–100 cm³ are a good choice for many refractories.

Besides adequate measuring volume other requirements are to be considered:

- Temperature during high temperature characterisation has to be well defined which is supported by using muffle fur-

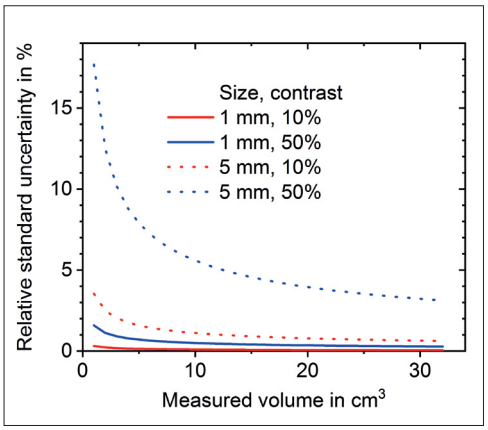

Fig. 1 Effect of measuring volume, characteristic size of constituting phases, and material contrast on variation of resulting material property

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naces and small feedthroughs to cold parts of the furnace.

- Furnace atmosphere should be controlled during the measurements. Otherwise, e.g. corrosion layers could be formed changing emissivity of the samples.
- In general, chemical or mechanical interaction of the measuring system with the sample should be avoided, since high temperature reactions can occur. Therefore, non-contact measuring methods are to be preferred.
- Since heating up of samples to a well-defined test temperature takes a lot of time, efficiency is significantly improved using multiple sample holders and high temperature sample change.
- In the same way measuring devices which can provide various material data simultaneously are to be preferred.

2 Customary measuring of thermal conductivity

Various measuring methods for thermal conductivity respectively thermal diffusivity exist. They are divided into stationary and transient methods. Stationary methods use a constant heat flow through the sample which is measured together with the corresponding temperature difference. With transient methods a time dependent temperature measurement is required and heat is applied either by single pulses or periodically. Heat is produced by resistance heating or by radiation from infrared lamps or lasers. Temperature is measured either by thermocouples, resistance thermowires or by pyrometers.

Highest measuring temperatures are achieved using the so called laser-flash method. The idea was introduced already in the 1970s when single pulses of infrared lamps were used instead of lasers to heat the front face of disk shaped samples [1]. From the temperature evolution at the rear side of the samples, thermal diffusivity is determined using a series expansion of Fourier's law for one dimensional heat flow [1]. Sample size was restricted to about 10 mm diameter and a few millimetres thickness since a homogenous heating of the entire sample surface was required and lateral heat losses had to be minimised to ensure one dimensional heat flow.

An improvement of the laser-flash technique towards high temperature measurement was presented in the 1990s. A first Thermooptical Measuring (TOM) device was developed which uses a CO_2 laser for heat production [2] instead of a customary Nd-YAG laser or an infrared lamp. By the extremely long wavelength of CO_2 lasers of 10,6 µm it was achieved that heat radiation was absorbed in a very thin layer at the samples surface. Correspondingly, a sensitive pyrometer was developed for temperature measurement at the sample's rear side working in the long infrared wavelength range [2]. By these measures a coating of the sample surface which is customarily applied could be avoided. This is an important advantage since coatings can react with the sample at high temperatures and they produce additional radiative heat transfer leading to measuring artefacts. Using the TOM device, laser-flash measurements up to temperatures of more

than 2000 °C could be successfully performed. A second improvement of the technique was introduced in 2006, when an inverse Finite Element (FE) simulation of heat flow was used for data evaluation instead of the serial expansion of one dimensional heat flow. This FE simulation considers three-dimensional heat flow allowing for larger samples of about 35 mm diameter and 15 mm thickness [3]. In the meantime also commercial laser-flash devices are offered with operating temperatures above 2000 °C [4]. However, with customary laser-flash techniques sample size is still too small for refractories. Laser-flash measurements are described in several standards, e.g. ASTM C518.

Stationary heat flow measurements are usually made using hot plate (ASTM C518, ASTM C177) or comparative cut bar methods (ASTM E1225). It was shown that they can be applied up to temperatures of 1200 °C [5]. However, handling of lateral heat losses becomes increasingly difficult and the correction for the temperature dependence of thermal conductivity is a challenge. So for high temperature measurements of large components hot wire (compare EN 993-14) or hot disk methods [6] are preferred. They basically use metallic wires which are embedded in the sample to produce local heat in addition to the homogeneous temperature field generated by the furnace. Then the temperature response is measured either by thermocouples placed close to the heat source or by the temperature dependence of the electrical resistance of the heating wire itself. Hot wire and hot disk methods can be used for large samples up to temperatures of about 1200 °C. A well-defined thermal contact between heating wire, thermocouples and sample is important and chemical reactions between metals and sample have to be avoided.

Another approach, based on the old Ångström method, was presented recently [7]. It avoids heating wires and thereby achieves higher temperatures. For that, the temperature of a furnace was varied periodically and the phase shift between furnace temperature and sample temperature was measured. Large samples can be used with this method if the frequency of the temperature oscillation is adapted to sample size and thermal diffusivity. Data evaluation is facilitated using highly symmetrical furnaces, muffles and samples, e.g. a cylindrical setup [7]. If temperature measurement is done by using a pyrometer which is focused on an internal surface through a hole in the sample, a non-contact arrangement is obtained which can be used up to very high temperatures.

3 Customary measuring of other thermal properties

Specific heat and density of refractories are required in thermal management because they concatenate transient and stationary heat flow as well as heat transfer between fluids and solids [compare (eq. 1)]. High temperature density can be determined from measurements of room temperature density, e.g. according to Archimedes principle, and measurement of thermal expansion. The latter is easily done by dilatometry. Although, measuring volume is small, with many customary dilatometers the accuracy is sufficient with respect to thermal management, since the change of density with temperature is of the order of only 1 %/1000 K.

However, specific heat shows a steeper increase and its measurement at high temperatures is difficult. Drop calorimeters can be

Fig. 2 Photograph of the thermooptical measuring device TOM_wave showing the upper part of the high temperature furnace, and – in the background – the CO_2 laser

used up to 2000 °C but their use is tedious since samples have to be heated to several temperature levels in individual runs. An alternative are Differential Scanning Calorimeters (DSC) but they can use only small samples with a mass between 10–100 mg. Large refractory samples can be milled and samples can be extracted by splitting the powder using sample dividers. Since specific heat is an additive material property it is in general not changed by the milling process. However, DSC becomes increasingly difficult at temperatures above 1000 °C due to radiative heat transfer. DSC and drop methods for measuring specific heat are described in EN 821-3.

Furthermore, determination of specific heat is reported in connection with laser-flash measurements. Here the laser-flash signal response of a reference sample with well-known specific heat is compared to that of the test specimen [8]. Yet, the above mentioned restrictions on sample size prevent use of these comparative methods for heterogeneous refractory materials. Finally, the hot disk method provides data on specific heat as well [6] but – as mentioned in the previous section – it is restricted to temperatures well below 1200 °C. So an efficient measurement of specific heat for high temperatures and large sample volumes is not available yet.

Fig. 3 Sample holder and muffle of TOM_wave

Besides specific heat and density also heat transfer by convection and radiation has to be considered in thermal management. The heat transfer between furnace and any solid component is described by:

$$\dot{Q} = A \cdot [\alpha \cdot (T_f - T_s) + \sigma \cdot \varepsilon \cdot (T_f^4 - T_s^4)] \quad \text{(eq. 2)}$$

where $\dot{Q}$ is the heat flow from the furnace to the solid component and A its surface area. α is the coefficient of heat transfer, σ the Stefan-Boltzmann constant and ε the hemispherical total emissivity. T_f and T_s are furnace temperature respectively solid temperature specified [K]. At temperatures above 1000 °C the contribution from convective heat transfer can usually be neglected and heat flow is controlled by emissivity ε. Often directional emissivity is measured instead of hemispherical emissivity.

Assuming that the solid is a Lambertian radiator, where the emitted intensity doesn't depend on the angle to the surface normal, allows for an easy conversion of the respective quantities [9], but often this condition is not fulfilled.

Spectral emissivity is measured most frequently using infrared spectrometers where a scaling is done based on the radiation of a reference sample [9]. Total emissivity can be obtained from spectral emissivity by integrating over a wide wavelength range including the major part of heat radiation. The reference sample is preferably a black body radiator which has the same temperature as the sample. Temperature control is an important issue in measuring emissivity since reference sample and sample should have exactly the same surface temperature which has to be measured accurately [9]. These constraints complicate high temperature measurements of directional emissivity at large sample areas. However, this would be essential for the heterogeneous surfaces of refractories.

4 New concept

To comply with the requirements for high temperature measurements of heterogeneous solids (compare first section), a novel thermooptical measuring device was designed (Fig. 2). It was named TOM_wave since excitation and detection of measuring signals are performed non-contact using electromagnetic and acoustic waves [10]. Disk shaped samples with diameters up to 35 mm and a thickness of up to 20 mm are used providing a sample volume of about 20 cm^3. A sample holder for five samples enables automatic sample change at high temperatures, so that a total measuring volume of 100 cm^3 can be achieved (Fig. 3). Samples are fixed by three pins at their lateral surfaces to minimise heat flow to the sample changer.

The upper pin is freely movable in the vertical direction to enable measuring of the coefficient of thermal expansion by an optical method which monitors the displacement of this pin relatively to the sample holder.

The sample holder is placed in a $MoSi_2$ heated muffle furnace with an alumina muffle of 260 mm diameter and 180 mm height providing homogenous wall temperatures (compare Fig. 3). Maximum temperature of the furnace is 1750 °C. All feedthroughs are smaller than 30 mm in diameter to avoid substantial heat losses from the sample to the cold parts of the furnace. Solid angles (view factors) from the sample to the feedthroughs are smaller than 0,005 steradian. The furnace is insulated by vacuum

moulds of mullite fibres with a thickness of 120 mm to obtain stable temperatures in the inside. Furnace atmosphere is controlled and also vacuum conditions can be applied. All relevant material data for heat transfer can be measured with TOM_wave in different setups which can be changed automatically when the furnace is at high temperature. This and the automatic sample changer provide high efficiency of the measurements.

Fig. 4 shows the setup of TOM_wave for laser-flash measurements. A CO_2 laser with a power between 100–500 W is used for heating the sample at one side. Spot size of the laser beam on the sample surface is typically 10 mm and laser intensity follows a top-hat profile during laser-flash measurements. The temperature evolution is measured by a special pyrometer (HgCdTe detector, wavelength range 6,5–9,0 µm) at the rear side of the sample. A second pyrometer is focused on the lateral sample surface to monitor heat flow perpendicular to the symmetry axis. This pyrometer is especially useful for measuring orthotropic thermal conductivity.

Fig. 5 shows the temperature measurement during a holding period of the furnace. Temperature variation is as low as ±0,14 K demonstrating superior thermal stability of the setup. At the end of the holding period a stepped temperature rise of 10 K was introduced which is required for temperature calibration of the pyrometers. The pyrometer response is also shown in Fig. 5. Temperature calibration has a measuring uncertainty as low as 0,4 %, which enables

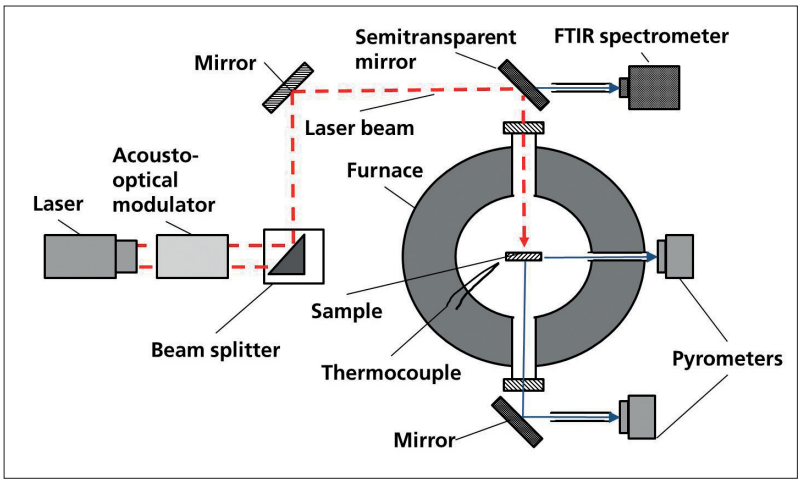

Fig. 4 Schematic sketch of TOM_wave together with the laser beam path during laser-flash measuring

an accurate determination of hemispherical total emittance. The pyrometer signals are interpreted by an inverse FE simulation of the three-dimensional heat flow within the disk shaped samples. Rotational symmetry of the sample enables a fast calculation of transient temperature fields and an iterative fitting of heat flow parameters. Fig. 6 shows the measured and the fitted temperature response of an alumina and a bauxite sample. The latter was extracted from a refractory brick B80. Good agreement between measurement and inverse simulation is obtained. The corresponding samples are shown in Fig. 7. The alumina sample was divided into smaller disk shaped samples which were measured in a customary laser-flash device (Netzsch LFA457) at the same temperature. Thermal diffusivity of the alumina sample was 1,343 mm²/s and 1,368 mm²/s measured in TOM_wave and the LFA457 respectively. The difference is within the range of results obtained in round robin tests with commercial laser flash devices (e.g. 3 % in [11]). This demonstrates that a successful upscaling of the laser-flash technique to larger sample volumes was achieved. The thermal diffusivity of the bauxite sample was 0,493 mm²/s. Five small samples of the same refractory material have been successively measured with the LFA457 with a mean thermal diffusivity of 0,48 mm²/s and a standard deviation of 0,05 mm²/s. On the one hand this confirms the good agreement of the two methods again and on the other hand the large standard deviation of the customary laser flash measurement illustrates the advantages of measuring large sample volumes.

Total hemispheric emissivity is obtained as well from this inverse fitting process of

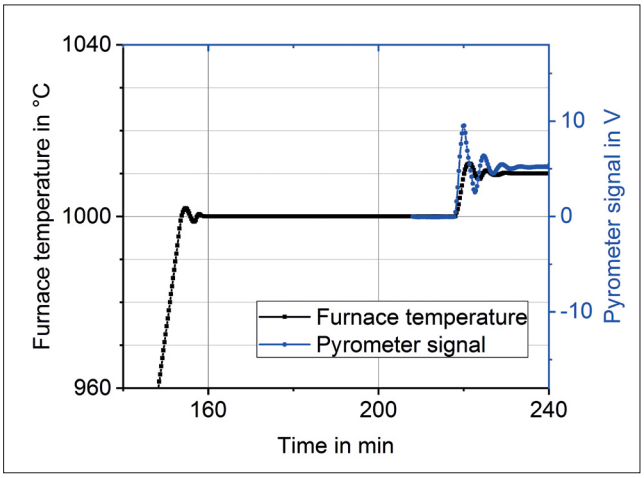

Fig. 5 Temperature profile during a holding period with subsequent step heating

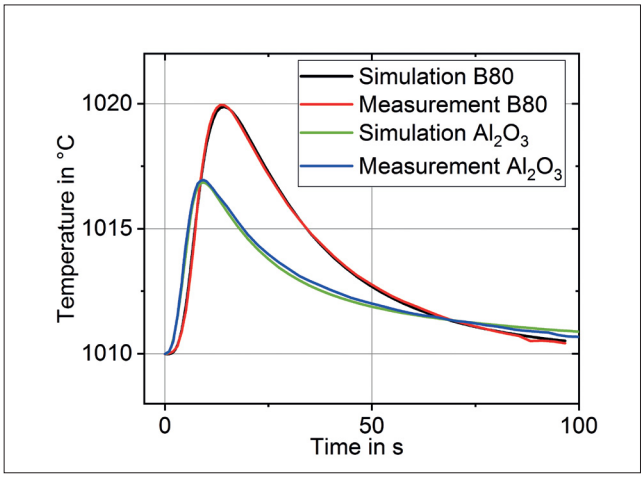

Fig. 6 Laser-flash measuring of an alumina and a bauxite (B80) sample: comparison of measured and simulated temperature curves

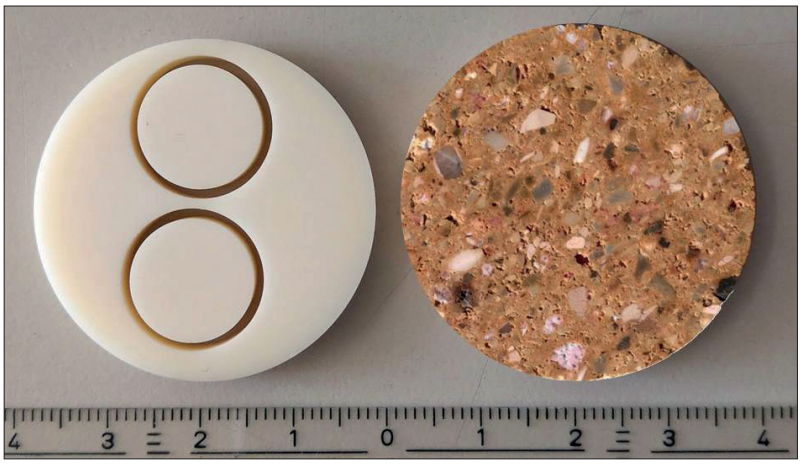

Fig. 7 Used samples: alumina disk with two smaller samples extracted for the comparison with LFA457 measurement (l.), and bauxite disk extracted from a refractory brick (r.)

the laser-flash signal (compare Fig. 6). It is 0,37 and 0,66 for the alumina and bauxite sample respectively. Since the entire sample surface contributes to the heat losses after laser-flash heating a large area is measured. This provides representative measurements with heterogeneous surfaces. On the other hand, a comparison with customary directional emissivity data can also be obtained. For that, a commercial FTIR spectrometer with a wavelength range from 0,9 µm to 28,6 µm is used (Bruker EM27-BO) at the furnace of TOM_wave (compare Fig. 4). At temperatures below 1000 °C vacuum can be applied during the measurement to avoid heat losses by convection [compare (eq. 2)]. A blackbody radiator is used as reference sample. Due to the high temperature homogeneity of the muffle, precise emissivity measurements can be obtained.

Specific heat can be directly extracted from the inverse simulation of the temperature evolution after laser-flash heating if the absorbed heat from the laser-flash is known. Pulse energy can be measured by the temperature evolution obtained after laser heating of a black body radiator and inverse FE simulation. In addition, the directional spectral emissivity of the sample is measured at the respective temperature. Alternatively, the comparative method [8] can be used for determination of specific

heat. Unlike its use described in [8], with TOM_wave no absorbing coatings have to be applied to ensure complete absorption of the laser light. Instead, directional spectral emissivity of sample and reference can be measured in exactly the same area which is heated by the laser (compare Fig. 4). Using TOM_wave, the main benefit with direct as well as comparative measurements of specific heat is that a large sample volume contributes to the measurement.

5 Concluding remarks

The thermooptical measuring device TOM_wave has been developed to enable an efficient high temperature measurement of heat transfer properties of solid materials. Sample volume is sufficiently large to provide representative measurements of heterogeneous materials like refractories. All relevant properties, i.e.: thermal diffusivity, specific heat, spectral and total emissivity and thermal expansion are measured at temperatures between room temperature and 1750 °C. Thermal conductivity is calculated from these data according to (eq. 1).

TOM_wave can also measure thermal shock behaviour of materials, which was already presented in a previous article in this journal [12]. Thermal shock resistance is an important property in life cycle assessments. Different from customary thermal shock testing a hot thermal shock is applied using the CO_2 laser for rapid heating of the samples. Crack formation is detected by four microphones which are installed at the furnace. By that thermal shock parameters can be investigated in a well-defined way which enables its use in FE simulations of service behaviour of high temperature components. Service life of high temperature components should be carefully considered in the development of new heating processes since it significantly contributes to their environmental footprint. Altogether, the high temperature measurement of thermal and mechanical material properties is an important contribution to a more purposeful design of high temperature processes.

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