Device for thermal conductivity measurement of exothermal material

Naprava za merjenje toplotne prevodnosti eksotermnega materiala

Grega Klančnik[1,] *, Urška Klančnik[2], Jožef Medved[1], Primož Mrvar[1]

[1]University of Ljubljana, Faculty of Natural Science and Engineering, Department of Materials and Metallurgy, Aškerčeva 12, SI-1000 Ljubljana, Slovenia
[2]University of Ljubljana, Faculty of Mathematics and Physics, Department of Techniques of Measurement in Physics, Jadranska 19, SI-1111 Ljubljana, Slovenia.

*Corresponding author. E-mail: grega.klancnik@ntf.uni-lj.si

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Abstract: Thermal conductivity is an important parameter, which helps us to design vital components in industry, where the right geometry (thickness) and material is needed.

The main role of measuring thermal conductivity is in improving the existing data bases with data for new materials. Usually unmeasured materials are those with intense reaction areas. Some are called exothermal or insulation materials. They are made from different exothermal materials which means, that the heat is liberated during an exothermal reaction. For exact determination of thermal conductivity more samples are usually required, especially if exothermal-insulation material is investigated. The samples are thermally very unstable. In this paper the solution is presented by using two methods in a single experiment with one sample.

It is also discussed which method is better for determining the thermal conductivity in the intense reaction area. It is interesting that one of the methods is used for absolutely steady state conditions, and the other one is a typical transient method. The measurements were obtained for the temperature interval from 100 °C to 900 °C.

Izvleček: Toplotna prevodnost je pomembna lastnost, ki nam omogoča lažje konstruiranje vitalnih delov v industriji, kjer sta potrebna pravilna geometrija (debelina) ter material.

Original scientific paper

Glavna vloga meritev toplotne prevodnosti je dopolnitev baz podatkov s podatki novih materialov. Navadno v bazah manjkajo podatki za materiale z intenzivnimi predeli reakcij. Taki primeri so eksotermni ali izolacijski materiali. Ti so sestavljeni iz različnih eksotermnih materialov, kar pomeni, da se toplota pri eksotermni reakciji sprošča. Za natančnejše določanje toplotne prevodnosti je treba opraviti večje število meritev, posebno pri preiskavah eksotermnih izloacijskih materialov. Vzorci so navadno toplotno nestabilni. V tem delu je predstavljena rešitev, ki zajema dve metodi z enim preizkusom ter z enim vzorcem.
V delu je prav tako nakazano, katera metoda je primernejša pri določanju toplotne prevodnosti v predelu intenzivnih reakcij. Zanimivo je, da ena metoda zajema popolnoma enakomerne pogoje, medtem ko druga metoda spremenljive. Meritve so bile izvedene v temperaturnem intervalu med 100 °C in 900 °C.

Key words: thermal conductivity, exothermal materials, differential thermal analysis
Ključne besede: toplotna prevodnost, eksotermni materiali, diferenčna termična analiza

INTRODUCTION

Thermal conductivity is a physical property with a well known definition: thermal conductivity is the property of a material that indicates its ability to conduct heat[1]. The Fourier thermal conduction equation states that the density of heat flux Φ/A is proportional to the temperature gradient[2]. Fourier law:

$$q = \frac{\Phi}{A} = -k \cdot gradT \qquad (1)$$

q – density of heat flux
Φ – heat flux
A – area
k – heat conductivity

Equation 1 can be written in a finite form as[1]:

$$\Phi = k \cdot A \frac{\Delta T}{\Delta L} \qquad (2)$$

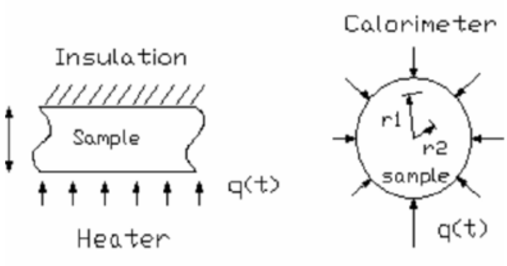
where Φ represents the heat flux trough a thickness ΔL on a surface A. Equation 2 is used for measuring a single thermal property at a time under steady state conditions. The calculated value of k is usually an average thermal conductivity in a specific temperature range. Using equation 2 can perform a significant error when thermocouples are used, because the location of thermocouples is not included. Instead the insertion of thermocouples can be made inside the upper cover of the sample and inside the heater plate to attain repeatedly. Without the cover the upper thermocouple could measure the temperature of the atmosphere instead of the sample, if the preparation of the measurement is not sufficient.

In the year 1974 Beck and Al-Araji[3] introduced a transient solution for the simple calculation of thermal conductivity. One of the advantages of this method is that the location of thermocouples is included in the calculation. Another advantage is a shorter time of measuring, but that also depends on the thickness of a sample and the quantity of heat added into the sample. In our calculations we used equation 3 for a flat plate:

$$k = \frac{LQ\left[(\frac{x_1}{L} - 1)^2 - (\frac{x_2}{L} - 1)^2 \right]}{2\int_0^\infty \left[T(x_1,t) - T(x_2,t) dT \right]} \qquad (3)$$

where x_1 and x_2 represent the location of the thermocouples, L is the thickness of the plate and Q the heat passing through the plate. Figure 1 shows a schematical drawing of a sample with a plate geometry and a solid cylinder.

From Figure 1 it is seen that with the same geometry of a sample, different approaches for heating can be made. If the calorimeter is used as a heater device and solid cylinder for the sample, no insulation is needed, because in a while the temperatures of the first (initial) and second (final) thermocouple will both approach the same value (Figure 2). In our experiment we also used a cylinder, but we did not use a calorimeter as a heater, but instead a separate heating device. In this case the calorimeter was used just for preserving the same working temperature (isothermal cell).



(a) (b)

Figure 1. Various geometries with one surface heated and the other insulated for: (a) Plate geometry and (b) without insulation for a solid cylinder[2]

The measurements were made on metals and insulation materials with both methods. If both methods were not used, several samples would have been necessary for determining the thermal conductivity constants. The right constant would be calculated by using an average of the recorded values[4].

EXPERIMENTAL WORK

In the present research a device for thermal conductivity measurement was designed. The temperature range for measuring is between 100 °C and 900 °C. The furnace had to be preheated, so when the heater was turned on, the heat flow was directed entirely into the sample. Otherwise the heater would heat the entire device, not only the sample, thus the heat losses created would not have been negligible. We used two type K thermocouples in our measurement, because of a good sensitivity. The thermocouples were connected to the measuring card from the National Instruments and the card to the personal computer. Signal Express 2.1 software was used for determining and converting the signal from the thermocouples. The measurement was started, when the temperatures from both thermocouples approached the same value. Measurements started when the initial and final temperature were in proximity to each other. The corresponding factor of thermal conductivity is at an average temperature $(T_{\text{Initial}} + T_{\text{Final}})/2$. Initial and final temperature rises till initial temperature closes to the final one. The whole measurement was terminated, when the temperature of the initial and final thermocouple stabilized[2].

Device for measuring thermal conductivity of exothermal materials

For measuring thermal conductivity at various temperatures, the furnace with capability of heating the sample up to 1000 °C was needed. The lower tunnel furnace with a sufficient and stable heating cone was used. This is needed to achieve a homogeneous temperature all over the sample, before starting the measurement. The heat flux was made with another heater, located under the sample. The heater was connected to the VDC PS3020 Lab laboratory power supply (0–30 V). The schematic presentation of the designed device is shown in Figure 2.

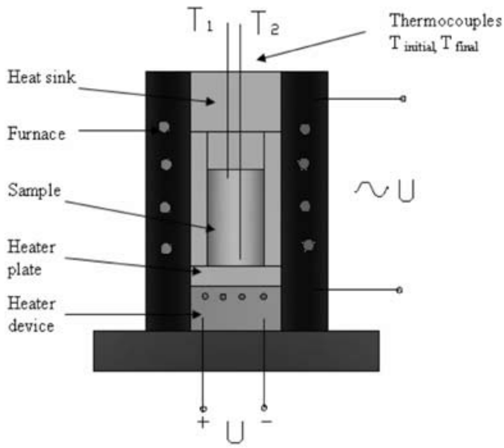


Figure 2. Device for thermal conductivity measurement – a schematic presentation

The construction of the device for thermal conductivity measurement is similar to the one described by J. FILLA[5, 6] specified for measuring thermal conductivity of ceramics.

The Heater

Kanthal wire was used in constructing the heater. The diameter of the wire was 0.5 mm, with a measured electrical resistance of 5.3 Ω/m. The wire was protected with corundum tubes. The wire was inserted on the top of the concrete grounding with good thermal and electrical resistance. The good thermal resistance was needed to maintain a one-dimensional heat flow. One of the options is discussed in papers by Beck and Al-Araji[2], where for the substrate two metal (copper) disks are used. The wire is placed in the middle of the two disks. Two samples are needed for this solution.

Heat sink

In this case the heat sink exists just for a small period of time in comparison to the whole measurement. During this period a linear temperature dependency appears, which is ideal for calculating the first average value of thermal conductivity according to Fourier. When the cooling system is switched of, the heat sink began to behave as an insulator. The time period must be long to minimize the effect of the first measurement. That is why the low current and voltage were used to get the temperature loop as long as possible.

The Measurement

The measurement of each sample started, when the initial and final temperatures were stable and in proximity. In this case the equilibrium of the device was reached. When the heater was turned on with an accurate electric power, the measurements began. The added heat into the sample was always between 7 W and 8 W, which is very low. With that, overheating of the sample was prevented and the linear rise of both temperatures was easier to achieve. Because of the low current and voltage used, a longer time was needed for the experiment, which was an advantage for determination of thermal conductivity using Fourier's law. For calculating constants using Fourier's equation was always done in the temperature region where the linear temperature dependence exists. The whole measurement was taken under air. The highest error appeared in metals, especially in aluminum, which was expected, because the temperature differences were less than 1 °C. An error can quickly appear if samples used are too short. Also, the value of added heat into the sample is also of importance.

Results and discussion

The complexity of the measurement will appear when exothermal material is examined. The starting (working) temperature must be determined in a way, that the whole exothermal reaction is measured and calculated according to both equations. The results for magnesium, calculated with Equations 2 and

Table 1. Thermal conductivities for magnesium

Temperature, T/°C	k/(W m^{-1} K^{-1}) Fourier	k/(W m^{-1} K^{-1}) Beck-Araji	k/(W m^{-1} K^{-1}) Average value	k/(W m^{-1} K^{-1}) Literature[7]	Error, E/%
94	137.359	137.359	137.359	154.34	11
140.18	137.552	137.079	137.315	153.24	10.3
234.58	135.084	135.435	135.259	150.98	10.4
284.16	133.630	134.122	133.876	149.78	10.6
299.38	132.906	133.028	132.967	149.41	11

3, are presented in Table 1. It is seen that a small variation appears between the Fourier and Beck-Araji method. The meausered thermal conductivities are done without any calibration. The calculated values were than used for calibration.

After calibration, a diagram for thermal conductivity for magnesium was made. A good compromise was made with the calculation of average values of thermal conductivity for both methods.

It is seen from Figure 4 that both methods are comparable in the temperature range from 100 °C to 400 °C. The difference between both methods under 100 °C and over 400 °C appears because a temperature difference formed between the starting temperatures. The temperature difference did not appear because of a non equilibrium state, but because the temperature homogeneity was not reached. One of the ways how to eliminate the starting temperature difference without building a new

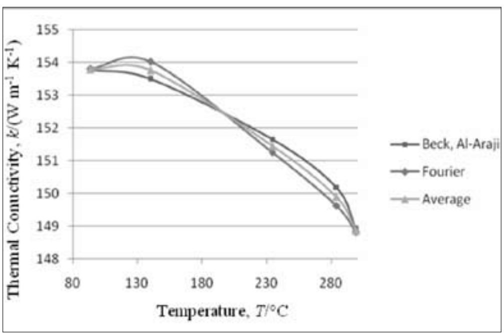


Figure 3. Thermal conductivity of magnesium calculated according to FOURIER[1] and BECK, AL-ARAJI[2]

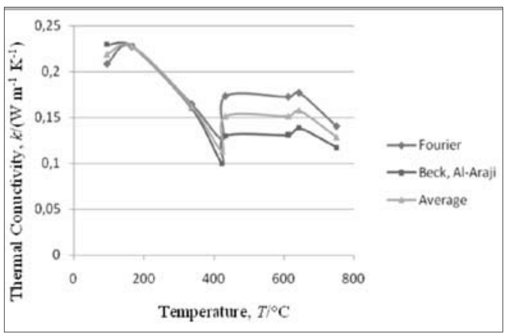


Figure 4. Thermal conductivity of insulation material

furnace is to make the samples smaller. With that solution the temperature homogeneity is easier to achieve. From Figure 4 it is seen that errors appear at higher temperatures, but nonetheless a clear tendency for both calculations is visible. Also noticeable in Figure 4 is the temperature drop at about 400 °C. This happens due to the internal heat source that arises as a result of the exothermal reaction occurring inside the sample itself.

With differential thermal analysis (*DTA*) the characteristic temperatures were obtained for the exothermal reaction. With that information, the starting temperature for measuring the thermal conductivity constant was determined. Differential thermal analysis was made on a DTA 701 Bähr device. The maximum temperature of heating was 1500 °C. The heating and cooling rate was 10 K/min under air. Alumina was used as a reference. From Figure 5a it is visible that between the temperature region between 313 °C and 439 °C the main exothermal peak appears. This is interesting for determination of thermal conductivity using both methods to see, if a comparison of both methods can be made. The starting temperature for the thermal conductivity measurement must always be chosen in such a way, that the reaction will appear in the steady state measurement first.

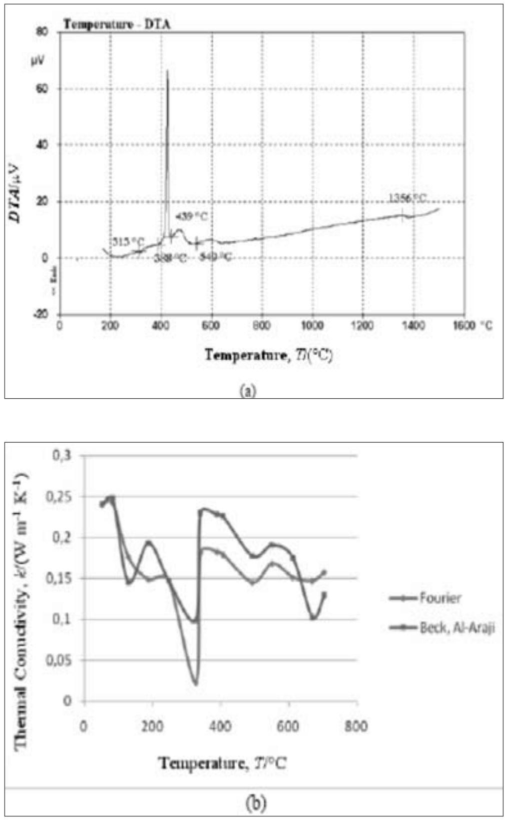


Figure 5. *DTA* heating curve (a) and thermal conductivity curve (b) for insulation material

The correspondent thermal conductivity diagram is presented in Figure 5b. It is seen that the determination of thermal conductivity according to both methods in the area of the exothermal reactions are similar, but not quite the same. Both methods showed a decrease in thermal conductivity when the exothermal reaction appeared, which indicates there was an internal heat source in the sample.

CONCLUSION

The basic idea of our work was to compare two different methods in a way that fewer samples are needed. In our calculations the Fourier and Beck, Al-Araji equations were used. The results showed that the combination of both methods can be used in the temperature range where no intense reactions are present. In the intense reaction range both methods showed similar tendency to lower constants. The equation according to Beck and Al-Araji has been shown as a good method for fast determination and estimation of thermal conductivity even for the intense reaction area. The best results were obtained under steady state conditions.

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