Determination of Experimental Conditions for Applying Hot Wire Method to Thermal Conductivity of Slag

Björn Glaser, Luyao Ma and Du Sichen*

Department of Materials Science and Engineering, Royal Institute of Technology, SE-10044 Stockholm, Sweden

Abstract: In order to apply the hot wire method for metallurgical slag at steelmaking temperature, a numerical model was developed, cold model experiments were conducted and test measurements using a high temperature experimental setup were carried out. To minimize natural convection and obtain more reliable measurements, the crucible diameter, the hot-wire diameter, the applied current, the position of the wire in the crucible and the cooling on the upper surface of the crucible were studied. Investigations on the sheathing material of the circuit exposed to the slag were also made. The hot wire resistivity was measured to make the thermal conductivity calculation more reliable. It was found that only certain materials were suitable for slag measurements depending on slag composition and temperature. The wire diameter also played a major role, because of the heat generation per surface area. The thermal conductivity should be derived from the values measured during the first seconds. In this initial stage, the effect of the natural convection as a function of the wire position in the crucible, the cooling on the top surface and the diameter of the crucible are negligible. A compromise has to be made in choosing the electrical current, since higher current results in higher sensitivity but at the same time in more natural convection.

Key words: Thermal conductivity, Molten slag, Transient hot wire method, Line source method

1. Introduction

Heat is transferred by conduction, convection and radiation. To be able to properly control the heat transfer in many engineering applications, accurate material data are necessary, where the thermal conductivity of a material is one of the most important properties. Thermophysical property values are mainly used in calculations and modelling. In order to get good results, accurate data are essential.

In relation to the steelmaking industry, the thermal conductivity plays a critical role in all different steps of the steel making process. In these processes, good understanding and control of heat transfer is beneficial to control the production process and achieve the final composition of the steel. Hence, thermal properties of liquid steel and slag are extremely important.

Measuring the thermal conductivity of slags at temperatures beyond 1773 K is exceedingly difficult for many reasons, as evidenced by the lack of data above this temperature for many common slag compositions. For high temperature thermal conductivity slag measurement it has been found that the transient hot wire method has highest accuracy and mainly used above 1773 K.

A lot of researchers have developed methods to measure the thermal conductivity of solids, liquids and gases. Still there are only a few which measures the thermal conductivity of slag at high temperatures. Susa and colleagues [1] determined the thermal conductivity of alkali silicate melts containing fluorides using the non-stationary hot wire method. The measurements were carried out between 1050 and 1550 K. It was found that the thermal conductivity of these silicates decreases with increasing temperature and that the additions of fluorides to the alkali silicate melts decrease the thermal conductivity of these melts. Some conclusions about the ionic bonds were made.

The thermal conductivity of the $CaO-Al_2O_3-SiO_2$ system was measured by Kang and Morita [2]. The non-stationary hot-wire method was used up to a temperature of 1873 K. The effects of slag composition and temperature on the thermal conductivity were investigated.

Xie and colleagues [3] developed a transient short hot-wire technique to determine the thermal conductivity and diffusivity of various materials such as liquids, gases and powders. In order to improve the accuracy, a hot-wire of only 10 mm long was used. A two-dimensional heat-conduction model including thermophysical properties, size of the hot-wire, insulation coating and sample was developed. It was found that the effect of natural convection is negligible in the experimental heating time range. An error of 2% for the thermal conductivity and 7% for thermal diffusivity could be estimated.

Boron-containing slag is commonly used for the blast furnace. Zhan at al. [4] measured the thermal conductivity of boron-containing slag up to 1773 K. The transient hot-wire method was applied. It was found out that the thermal conductivity decreased with increasing temperature. The effect of different slag compositions at thermal conductivity was studied.

Nagata, Susa and Goto [5] investigated the thermal conductivity of synthetic slags and slag samples taken from the blast furnace. The synthetic slag systems studied were $CaO-SiO_2-Al_2O_3$, $CaO-SiO_2-Fe_2O_3$, $Na_2O-SiO_2-CaF_2$ and $CaF_2-CaO-SiO_2-Al_2O_3$. The thermal conductivity of the slag was measured in the temperature range 373 K - 1773 K, both in the solid and liquid. An empirical model for thermal conductivities of slags at high temperatures was developed.

The thermal conductivity of electrically conducting liquids was measured by Joseph and colleagues [6]. As the measurement system, the transient hot-wire method was used. A glass capillary filled with mercury was used as an insulated hot wire. The system was used up to 493 K.

The main focus of the present work is to determine the conditions for using the transient hot-wire method for slag systems. To reach this goal, a numerical model is to be developed and cold model experiments are to be conducted to verify the model predictions. Finally, test measurements using of a high temperature experimental setup are to be made to examine the reproducibility of the technique and the suitability of refractory materials.

2. Experimental Methods

The experimental methods for thermal conductivity measurements can be divided in three main groups: steady-state, non-steady state, and transient methods. In the case of steady-state methods the temperature is a function of spatial coordinates, but independent of time. In non-steady state methods, the temperature varies with time as well as position. Transient methods refer to systems where the temperature distributions at any point of the system vary continuously with time.

Steady-state techniques include the linear heat flow method and the radial heat flow method. In both methods the temperature profile across the specimen is determined by thermocouples. If these techniques are used for measurements of liquids, significant errors can occur from convective heat transfer.

The radial wave (RW) and the modulated beam methods are examples of non-steady techniques. In the radial wave method, the crucible is placed in the isothermal zone of a furnace and thermocouples are located on the walls and along the geometric axis of the crucible. The outside wall of the crucible is then subjected to a variation of temperature and the change in temperature of the central thermocouple is monitored. The measured thermal diffusivity values can contain errors caused by heat transfer due to convection and radiation. A laser beam which produces a periodic variation in temperature of constant frequency is used in the modulated beam technique. The used specimen is in form of a disk, where the front is heated with the laser beam. The phase shift between this input and the signal from a temperature sensor in contact with the back face is determined. Errors for measurements in liquids can be caused by heat transfer through convection.

The laser pulse (LP) and the line source (LS) technique produce a temperature-time curve by input of energy. The curve is monitored during the short time period of the experiment. These techniques are known as transient methods. A more detailed explanation of the different systems can be found in the literature. [7]

The line source (LS) method, also known as the transient hot wire method, was chosen as the one which is most suitable for high temperature experiments for two main reasons: First it is thought that the heat transfer by radiation is very low and therefore has little influence on the measured conductivity value. Secondly, any convection occurring results in a departure from linearity and can be excluded by using only the straight part of the temperature-time curve.

3. Principle of the line source (LS) method

The mathematical background of the line source method is well-established and can be described as follows. The heat generated by hot wire mainly travels through the sample by conduction as described in Eq. 1 where

$$Q = -k\frac{dT}{dx} \tag{1}$$

Q is the heat generation rate per unit length and dT/dx the temperature gradient along the direction of the heat flux, (here only x direction is considered).

The thermal conductivity k is related to the thermal diffusivity α, the heat capacity c_p and the density ρ by the following equation (Eq. 2):

$$k = \alpha C_p \rho \tag{2}$$

In numerical analysis, the sample is considered as a semi-infinite cylindrical system, where the hot wire is considered to be parallel to the z-axis. Therefore the energy equation can be written as given in Eq. 3.

$$\rho C_p\left(\frac{\partial T}{\partial t} + v\nabla T\right) = k\nabla^2 T + Q \tag{3}$$

The temperature increase of the wire ΔT can be obtained from Eq. 4 where

$$\Delta T = \frac{Q}{4\pi k}\ln\frac{4\alpha t}{r^2 C} = \frac{Q}{4\pi k}(\ln t + A) \quad (4)$$

t is the time, *r* is the radius of the hot wire, *C* = 1.7821 (the exponential of the Euler constant) and *A* is a calculated constant [8]. The thermal conductivity *k* can be calculated with use of Eq. 5.

$$k = \frac{Q}{4\pi} \Big/ \frac{dT}{d\ln t} \quad (5)$$

In the experiments, constant current is applied to the hot wire to generate heat. The voltage increase *V* of the hot wire is logged over time *t*. The heat generation rate per unit length *Q* and the relationship between *dV/dlnt* and *dT/dlnt* can be obtained respectively along with the resistance equation of the hot wire. The resistance equation for the cold model calculations is given in Eq. 6 and the one for the high temperature calculations in Eq. 7 where

$$R_T = R_{ref}\left[1 + \alpha\left(T - T_{ref}\right)\right] \quad (6)$$

$$R_T = R_{ref}\left[1 + \alpha T + \beta T^2\right] \quad (7)$$

R_T is the resistance at certain temperature and R_{ref} is resistance at reference temperature.

In calculations for cold model, only the first order temperature coefficient of resistance, *α*, of the hot wire is used. Since the relationship between resistivity and temperature difference at low temperatures is linear. For high temperature thermal conductivity calculations the first order and the second order temperature coefficient of resistance, *α* and *β*, are used, since the relationship between resistivity and temperature difference at high temperatures is non-linear.

The heat generation rate for both calculations can be determined using Eq. 8 where

$$Q = \frac{I^2 R_T}{l_{wire}} \quad (8)$$

I is the current used in measurements and l_{wire} is the length of the hot wire. Eq. 9 for cold model calculations and Eq. 10 for high temperature calculations shows the relation to the measured voltage of the hot-wire expressed with resistance respectively.

$$V = IR_{ref}\left[1 + \alpha\left(T - T_{ref}\right)\right] \quad (9)$$

$$V = IR_{ref}\left[1 + \alpha(T - T_{ref}) + \beta(T - T_{ref})^2\right] \quad (10)$$

Eq. 11 and Eq. 12 are derived from Eq. 9 and Eq. 10 respectively. The equations show the relationship between *dV/dlnt* and *dT/dlnt* for cold model calculations and high temperature calculations, where $\Delta T = T - T_{ref}$ where

$$\frac{dV}{d\ln t} = I\rho_{ref}\frac{l_{wire}}{\pi r^2}\alpha\frac{d(\Delta T)}{d\ln t} \quad (11)$$

$$\frac{dV}{d\ln t} = I\rho_{ref}\frac{l_{wire}}{\pi r^2}(\alpha + I\beta\Delta T)\frac{d(\Delta T)}{d\ln t} \quad (12)$$

ρ_{ref} is the resistivity of the hot wire at reference temperature.

The above equations show the relationship between heat generation rate per unit length *Q*, *dT/dlnt* and *dV/lnt*. But to get *dV/dlnt* from the curve *V/lnt* is not very straightforward.

In the beginning of the measurements, the current is not very stable, so the relationship between *V* and *lnt* is not linear. In the later stage of the measurements, the natural convection in the liquid sample has already started and cannot be ignored. The natural convection also causes a deviation from the linear relationship. Only the slope *dV/dlnt* of the linear part of the curve can be used for the calculation of the thermal conductivity value. An example of the curves in low and high temperature measurements is given in Figure 1. According to the experiments, the linear part of the curve in the cold model experiment is usually between 2 and 4 seconds of measuring time.

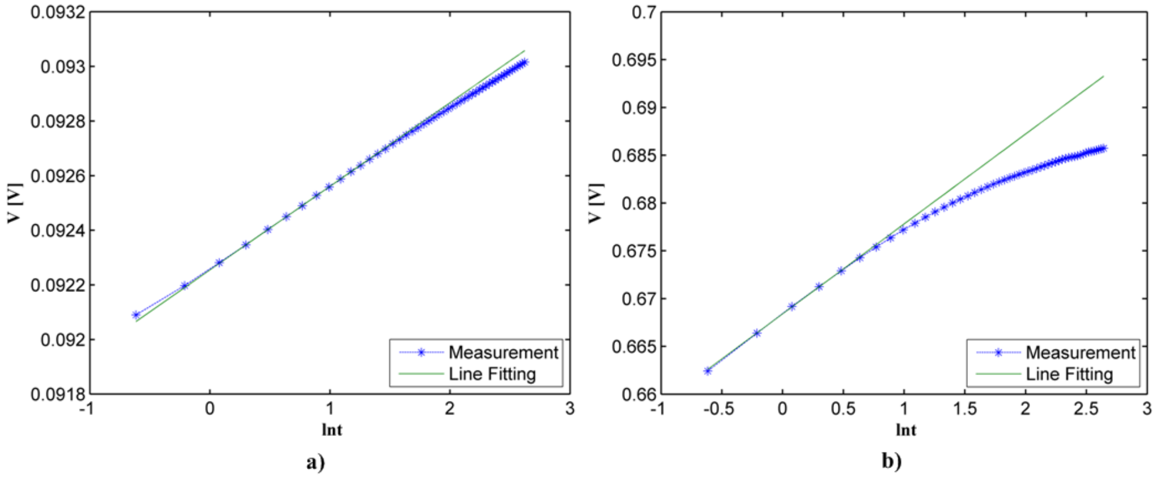


Fig. 1 Example of the line fitting a) low temperature measurement b) high temperature measurement.

For the high temperature calculations, the straight part is usually between 1 and 4 seconds of measuring time. ΔT in the term $I\beta\Delta T$ in Eq. 12 is the temperature increase of the straight part of the curve, where ΔT is calculated from Eq. 13, in which ΔV is the voltage increase of the straight part.

$$\Delta T = \frac{\left\lfloor -\alpha + (\alpha^2 + 4\beta\Delta V)^{0.5} \right\rfloor}{2\beta} \tag{13}$$

4. Mathematical modelling

To evaluate the use of the hot wire method for high temperature applications a mathematical model was developed using COMSOL Multiphysics 4.2a. The purpose of the modelling effort was to find the optimal dimensions of the crucible, wire thickness and wire position in order to minimize natural convection. Calculations with different dimensions of the crucible/wire and wire positions were carried out. The physical properties of the liquid and the wire used in the calculation were the same as used in cold model experiments (described in the following section), where the used liquid was Rhodosil Silicon Oil 550 and the hot wire was made of Pt-10%Rh and Pt-6%Rh (Table 1). The resistivity and the first order coefficient of resistance of the Pt-10%Rh and Pt6%Rh wire is given in Table 2. The resistivity and the first order coefficient of resistance were calculated from the cold model resistivity measurements.

Table 1 Material and physical properties used in COMSOL modelling.

Material	*Density (ρ) [kg/m³]*	*Thermal Conductivity (k) [W/(m*K)]*	*Heat Capacity (Cp) [J/(kg*K)]*	*Dynamic Viscosity (μ) [kg/(m*s)]*
Rhodosil Silicon Oil 550 [9]	1070	0.146	1506.24	0.127
Pt-10%Rh Wire [10,11]	19970	37.7	140	-
Pt-6%Rh Wire [10,11]	20550	45.0	137	-

Table 2 Wire resistivities and temperature coefficient of resistance used in calculations for cold model measurements.

Wire Material	*Wire Diameter*	*Temperature*	*Wire Resistivity*	*1st Order Temperature Coefficient of Resistance*
	[mm]	*[K]*	*[Ω*m]*	*[Ω/Ω*K]*
Pt-10%Rh	0.35	303	1.91988*10^{-7}	1.4567*10^{-3}
Pt-6%Rh	0.25	303	2.25995*10^{-7}	1.8805*10^{-3}

4.1 Simplifications and assumptions

The following simplifications and assumptions were made.

- Only the liquid domain was modelled, since only the effect of the natural convection was of interest (Figure 2a).
- Only the effect of the hot wire heated by joule heating was taken into consideration (Figure 2a). The other parts of the circuit were neglected.
- The heat transfer to the surroundings was neglected, since only the heat transfer in the liquid caused by the heating of the wire is of interest. The heat transfer from the top of the crucible, however, was taken in consideration.

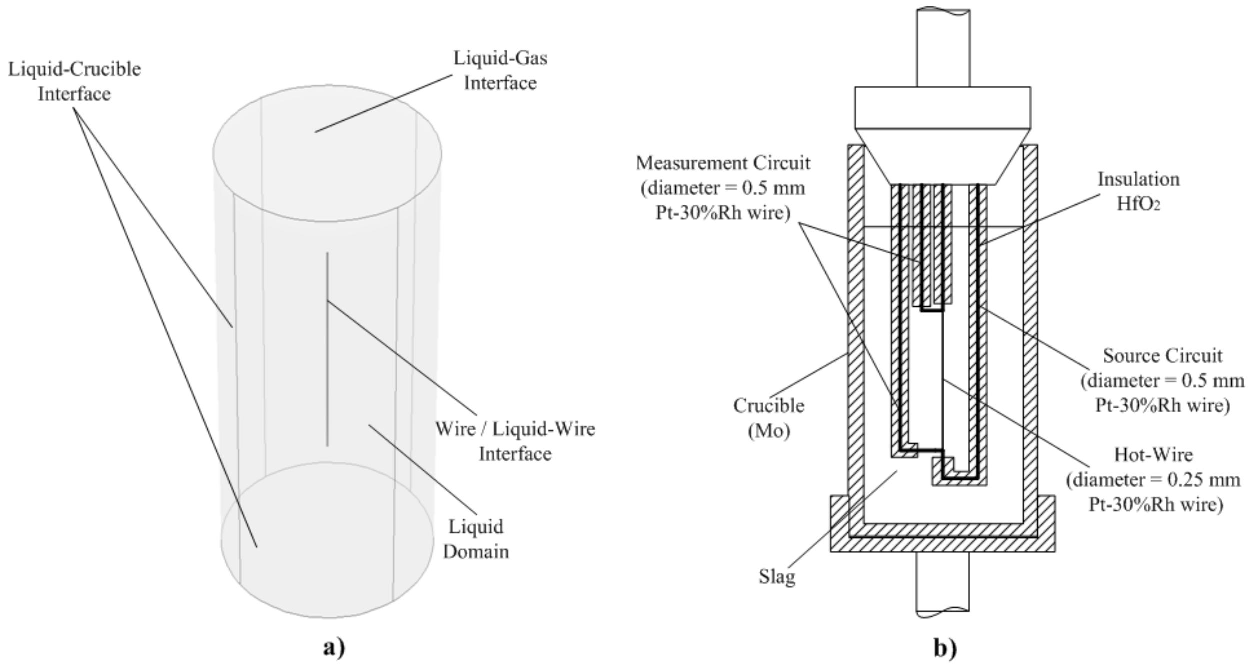


Fig. 2 Sketch of a) modelling domain b) hot wire circuit.

4.2 The domain of the model

In order to simulate the hot wire measurement process the turbulent flow application and the joule heating application in COMSOL were used. To study the fluid flow driven by buoyancy forces due to the density difference, a three dimensional model was constructed. The continuity equations, momentum equations and energy conservation equations were solved simultaneously.

4.2.1 Boundary conditions

At the liquid-crucible interface:

For the turbulent flow application, the interface between liquid and crucible was set to wall function for turbulent flow in order to describe the thin region near the wall with high gradients in the flow variables [12]. For the joule heating application the boundary was set to thermal insulation. The heat loss through the crucible wall and to the surroundings is not taken into consideration.

At the liquid-gas interface:

For the joule heating application the boundary was set to convective cooling in order to simulate the heat transfer to the surroundings, where the built-in COMSOL heat transfer coefficient for air was used and the external temperature was set to 298 K. To describe the free surface with no fluid penetration and vanishing shear stresses [12], the boundary was set to symmetry in the turbulent flow application.

At the liquid-wire interface:

In the turbulent flow application the boundary was set to wall function. For the joule heating application it was assumed the heat flux is continuous across the boundary in each direction.

4.2.2 Initial conditions

The initial temperature of the liquid and the wire was set to 303 K as used in the cold model experiments. The initial velocity of the liquid was set to zero.

4.3 Computing

For calculation the commercial software package COMSOL Multiphysics 4.2a was used. The mesh was set as a compromise between convergence and memory requirement and can slightly differ within the calculations. The default solver provided by the software package was used.

5. Measurements

To improve the accuracy and functionality of the hot wire method at high temperatures, several cold model measurements at 303 K where carried out. For better understanding and proof of reproducibility the conductivity of a certain slag was measured at high temperatures. A sketch of the setup used at room temperature is given in Figure 3a. The setup used for the high temperature measurements is shown in Figure 3b.

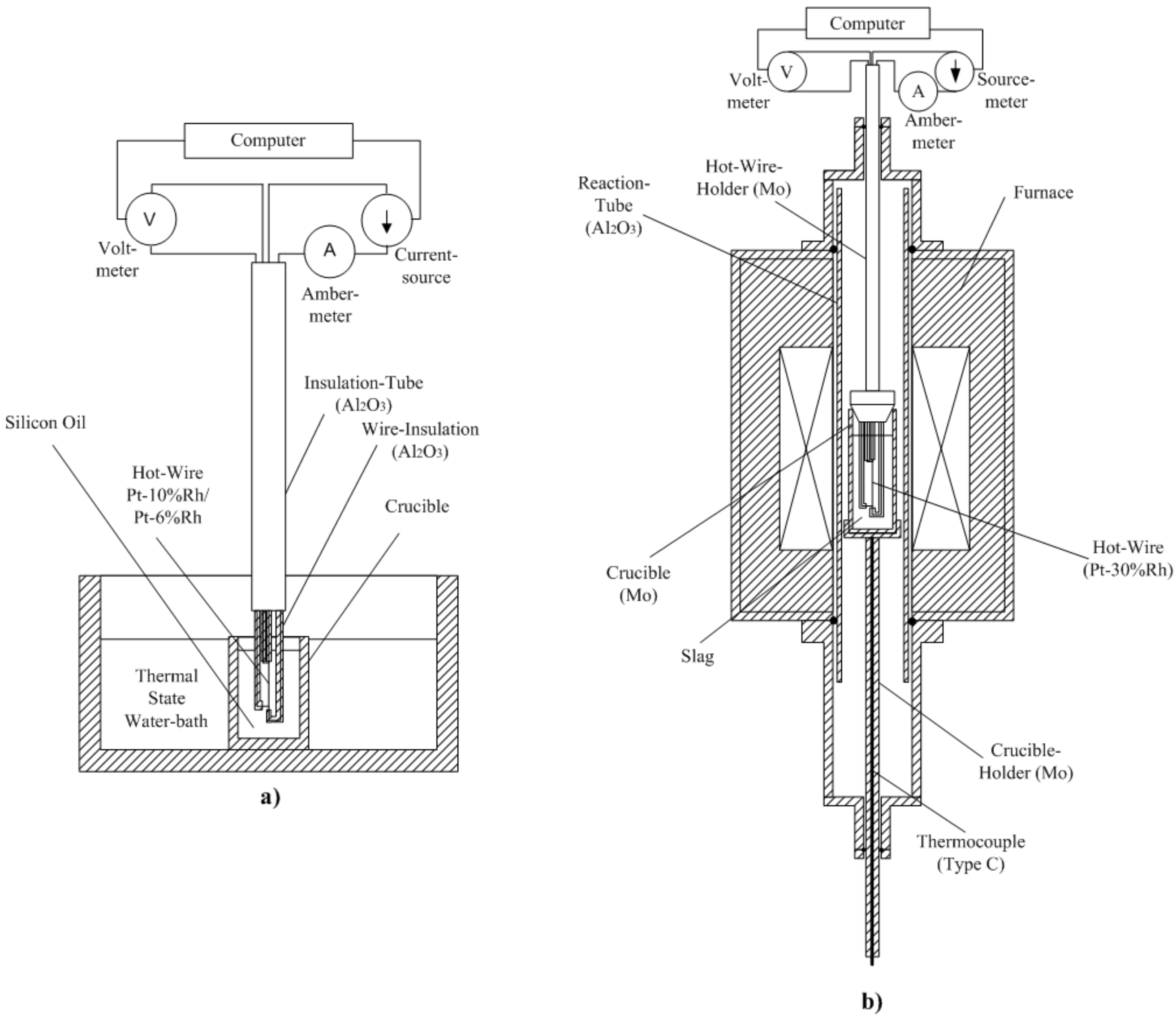


Fig. 3 Sketch of the a) low temperature experimental setup b) high temperature experimental setup.

5.1 Measurements at room temperature

The measurements at room temperature were carried out using a thermal state water bath. A sketch of the setup is given in Figure 3a. The measurement circuit contains the hot wire, extension wires, a source meter Agilent 66332A, and two multimeters Fluke 45/ Agilent-HP 34401A. The voltage change over time of the hot wire was measured and logged using the Agilent-HP 34401A. The source meter Agilent 66332A and the multimeter Agilent-HP 34401A were connected to a computer. The logging time between the values was 0.27 s and the total measurement time was 30 s. The whole measurement circuit (excluding the exposed hot wire) was sheathed in alumina tubes to avoid accidental contact between the wires and for insulation to minimize heat flux to the liquid. For the hot wire, Pt-10%Rh with a diameter of 0.35 mm and Pt-6%Rh with a diameter of 0.25 mm wire material was used. The modelling fluid was Rhodosil Silicon

Oil 550, which was chosen because of its similar viscosity to slag. The physical properties of the modelling fluid are given in Table 1.

The experiments were performed in the following manner. The thermal state water bath was heated up to 303 K. The crucible with the silicon oil was placed in the water bath for several hours to ensure the silicon oil had an exact temperature of 303 K. The hot wire was placed inside the crucible 10 minutes before measurement to make sure silicon oil was completely still after the movement of the wire. The measurement was carried out for 30 s. After the measurement the wire was removed from the liquid for 10 minutes for cool down to eliminate residual heat left which could influence the next measurement. All the measurements where performed in this way.

To be able to calculate the thermal conductivity, the resistivity and the temperature coefficient of resistance of the wire need to be known. Therefore, resistivity measurements for the Pt-10%Rh and Pt-6%Rh wire were performed in the temperature range 298 K - 313 K at every 5 K. The procedure of the resistivity measurements were as follows:

1. The water bath was heated up to 298 K.
2. The crucible with the liquid was placed in the water bath and kept there for several hours to make sure the silicon oil had an even temperature of 298 K.
3. The hot wire was placed inside the liquid exactly in the centre of the crucible and held there for 10 minutes to make sure the liquid was completely still.
4. To perform the measurements the source current was set to 0.2 A. For this kind of resistivity measurements the current needs to be kept as low as possible to avoid heating up of the wire through joule heating and therefore changes in resistivity. The measured voltage needs to become stable after few seconds. A current of 0.2 A was used in literature [13] to perform the measurements. At the time when the current was provided, the voltage change of the hot wire was logged over time.
5. The measured voltage became constant already after 1 s of measurement time. To avoid uncertainties the voltage was measured for 30 s.
6. After the measurement, the wire was taken out from the liquid for cool down to make sure there is no residual heat left which will affect the next measurement.
7. The same measurement procedure described above was applied for all temperatures in the range 298 K - 313 K.

The calculated resistivity curve for the Pt-10%Rh wire is given in Figure 4a. Figure 4a shows the resistivity curve as a function of ΔT. The resistivity ρ_T at each temperature can be calculated from the measured voltage with use of Eq. 14,

$$\rho_T = \frac{V_{stable}}{I} * \frac{\pi\left(\frac{d_{wire}}{2}\right)^2}{l_{wire}} \tag{14}$$

where I is the supplied current, V_{stable} is the measured voltage, d_{wire} the diameter, l_{wire} the length and A_{wire} the cross section area of the hot wire.

The figure also shows the line fitting curve. From the line fitting curve the resistivity ρ_T of the wire at each measured temperature and the first order coefficient of resistance α can be derived. The resistivity and the first order coefficient of resistance of the Pt-10%Rh and Pt-6%Rh wire is given in Table 2.

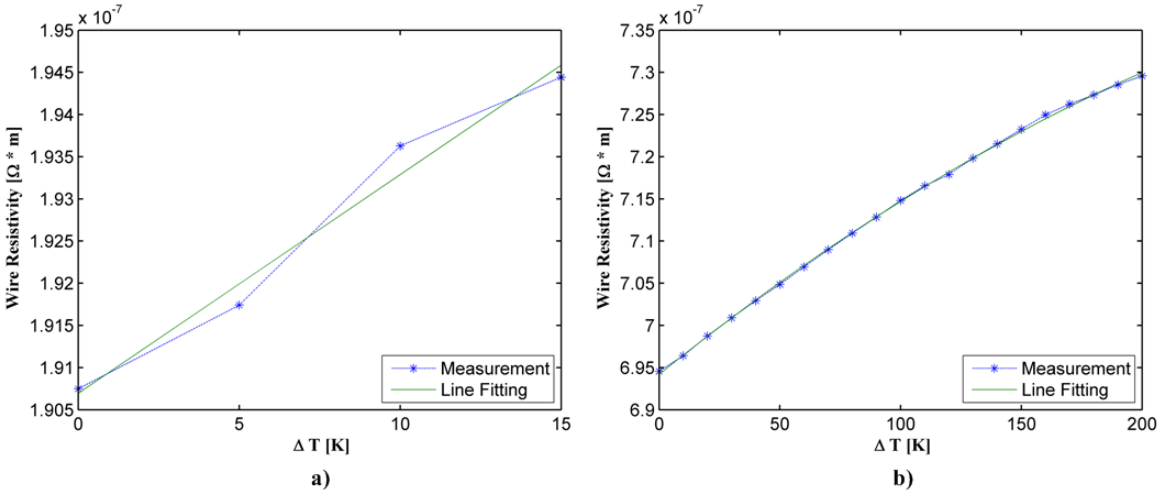


Fig. 4 Wire resistivity curve a) cold model, wire diameter 0.35mm b) high temperature, wire diameter 0.25mm.

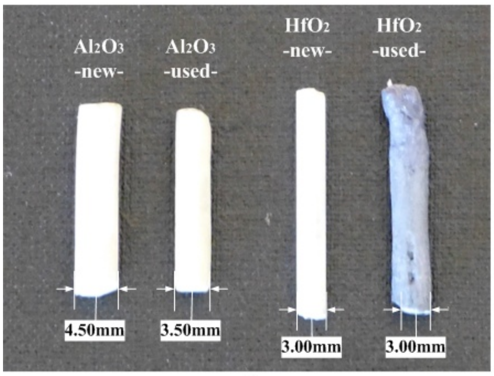
5.2 Measurements at high temperature

A sketch of the setup used in high temperature thermal conductivity measurements is given in Figure 3b. The setup was constructed to give the best performance for high temperature thermal conductivity measurements in a protective atmosphere. A furnace equipped with super kanthal heating elements up to 2023 K was used. The reaction tube, made of alumina, was sealed on the ends to avoid infiltration by oxygen. Argon was used as protection gas. To avoid cooling from the surface and therefore natural convection the crucible was placed with the slag surface in the hottest part of the furnace. Prior to the experiments the exact temperature profile of the furnace was measured.

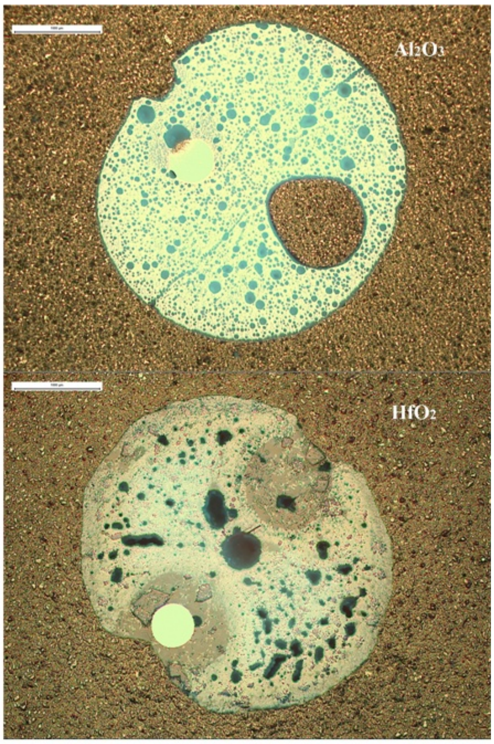
The crucibles, made of molybdenum, have an inner diameter of 35 mm and a height of 100 mm. The crucible dimensions were chosen based on the simulations and the cold model experiments, where it was found that the effect of convention in a crucible with smaller diameter is negligible. An inner diameter of 35 mm was chosen as a compromise between setup construction, measuring procedure and minimized convection.

The length of the hot wire was chosen to be 30mm because of the possibility to manufacture a proper hot-wire measurement circuit. The hot wire holder (Figure 3b) made of molybdenum was constructed in a way that the hot wire could be placed inside the crucible and taken out from the liquid slag without risk of infiltration by oxygen. The lower part of the wire holder (Figure 2b) was made in a conical shape to make sure the hot wire was centred in the crucible. The extension wires made of molybdenum placed inside the hot-wire holder were insulated with an alumina sheath to avoid electrical contact between wire and hot-wire holder. The wires of the circuit shown in Figure 2b which were submerged in the melt were made of Pt-30%Rh wire with an diameter of 0.5 mm. Only the hot wire which was used as heater and sensor was made of Pt-30%Rh wire with a diameter of 0.25 mm. Only the hot wire was directly exposed to the melt. All other wires of the circuit that came into contact with the liquid slag were sheathed in hafnia tubes to avoid extraneous heat flux to the surrounding and which could negatively influence the measurement results.

The temperature in the reaction tube was controlled by a type C thermocouple, mounted in the crucible holder made of molybdenum, directly positioned under the crucible as shown in Figure 3b. Hafnia (HfO_2) was chosen instead of more commonly used hard-fired alumina since it does not dissolve appreciably in the slag composition that was studied; Figure 5 shows the difference in dissolution between the two materials. Figure 5a shows the diameter of the alumina and hafnia cover tubes before and after measurements, where the hard-fired alumina tube was used for five and the hafnia tube for more than 60 measurements. The diameter of the hard-fired alumina tube is reduced after five measurements from 4.5 mm to 3.5 mm. However the hafnia tube does not show any reduction in diameter. From Figure 5a it can be seen that the hafnia tube is only slightly covered by slag. Figure 5b shows the cross section of the used hard-fired alumina tube and the used hafnia tube with use of an optical microscope. Here it can be seen in detail that some regions of the hard-fired alumina tube already dissolved in slag. The figure showing of the hafnia tube reveals only some penetration of the slag on the surface. This confirms that there is essentially no reaction between slag and HfO_2.



a)



a)

Fig. 5 Insulation sheaths a) comparison of the diameters between new and used insulation sheaths b) cross-section of the used sheaths.

By measuring and calculating the resistivity at the measured temperature and the first and second-order temperature coefficient of resistance of the hot wire, each wire material could be used for the hot wire measurements. However for high temperature thermal conductivity measurements of slag the materials to use are limited, since firstly the material has to withstand the high temperatures, and secondly the material has to be resistant to corrosion by the aggressive melt. It was found by experiment that pure platinum wire dissolves very rapidly in high silicate slags (more than 40% SiO_2) a pure platinum wire of 0.5 mm wire is dissolved in seconds. This makes it impossible to get reasonable results for this short period. Another factor is that the melting temperature of pure platinum is 2042 K and during the measurements at the 1873 K and 1923 K the pure platinum becomes very soft, which makes it impossible to place the wire for suitable measurements inside the melt. For the present measurements Pt-30%Rh wire is used because of its higher melting point (around 2200 K) and it was found that the higher the amount of rhodium in the wire the longer the wire can withstand SiO_2. This means a higher amount of rhodium in the wire or a pure rhodium wire would be more suitable for measuring the thermal conductivity in high silicate slags (more than 40% SiO_2).

The slag composition used for high temperature test measurements was 40%CaO-30%Al_2O_3-18%SiO_2-12%MgO. The slag composition is given in weight percentage. The conductivity of the slag was measured at 1773 K, 1823 K, 1873 K and 1923 K. In the first measurement cycle the conductivity was measured from the lowest to the highest temperature, whereas in the second cycle from the highest to the lowest temperature. At each temperature three measurements were performed. The two measurement cycles were completely independent of each other.

The chosen hot-wire diameter used in these experiments is 0.25 mm. From the cold model experiments we found that the wire diameter is a very important factor in the overall measurement process. We also know that too small diameter of the wire leads to convection which could deleteriously influence the measurements already from the start of the measurement.

The current was set to 1.5 A as suggested in literature[2]. From the cold model experiments it was discovered that the choice of the circuit amperage is in fact less important than the wire diameter. Therefore 1.5 A seems to be a suitable current level for the measurements.

The measurement instruments for the high-temperature measurements were the same as used in low-temperature measurements. The voltage increase of the hot wire was logged every 0.27 s. The measurements where performed in the following way. The crucible with slag was placed in the furnace with the top surface of the slag at the hottest point. The furnace was heated up to the temperature which was suitable for the measurement and kept there for at least one hour to make sure the slag had a uniform temperature distribution. The hot wire was submerged in melt for one minute before actual measurement to make sure the wire has the same temperature as the slag. The measurement was carried out for 15 s. After the measurement the wire was taken out of the melt for 15 minutes to eliminate residual heat left which would affect the next measurement. All the measurements were performed in this manner.

As already described above for the cold model experiments, in order to calculate the thermal conductivity it is necessary to measure the resistivity of the wire, where the temperature coefficient of resistance of the hot wire can be calculated. The resistivity measurement for the high temperature setup was performed as follows. The hot wire was placed in the hot zone of the furnace. The furnace was heated up to 1923 K. The measurements were performed in the same way as described for the low temperature experiments. The same current from 0.2 A was used. The calculated resistivity curve for the Pt-30%Rh wire is given in Figure 4b. Figure 4b shows the resistivity curve as a function of ΔT. The figure also shows the line fitting curve. From the line fitting curve the resistivity ρ_T of the wire at 1773 K, 1823K, 1873 K and 1923 K and the first and second order coefficient of resistance α and β can be derived (Table 3). The resistivity ρ_T can be calculated from the voltage measurements with use of Eq. 15

$$\rho_T = \frac{V_{stable}}{I} * \frac{\pi\left(\frac{d_{wire}}{2}\right)^2}{l_{wire}} \tag{15}$$

where I is the supplied current, V_{stable} is the measured voltage, d_{wire} the diameter, l_{wire} the length and A_{wire} the cross section of the hot wire.

Table 3 Wire resistivities and temperature coefficient of resistance used in calculations for high temperature measurements (Pt-30%Rh, 0.25 mm diameter and 30 mm long).

Temperature [K]	*Wire Resistivity* [Ω*m]	*1st Order Temperature Coefficient of Resistance* [Ω/Ω*K]	*2nd Order Temperature Coefficient of Resistance* [Ω/Ω*K]
1773	$7.0509*10^{-7}$	$3.3268*10^{-4}$	$-3.7418*10^{-7}$
1823	$7.1469*10^{-7}$	$3.3268*10^{-4}$	$-3.7418*10^{-7}$
1873	$7.2299*10^{-7}$	$3.3268*10^{-4}$	$-3.7418*10^{-7}$
1923	$7.3000*10^{-7}$	$3.3268*10^{-4}$	$-3.7418*10^{-7}$

6. Results

6.1 Modelling results

Figure 6 shows the velocity field caused by natural convection as a function of the crucible diameter at 15 s simulation time. For all the different crucible diameters the velocity after 15 s simulation time is very low, where the highest maximum velocity is 0.24 mm/s in the crucible with the largest diameter 8.81 mm. The lowest maximum velocity 0.21 mm/s is related to the crucible with the smallest diameter 3.8 mm. Thus, it is evident that the larger the diameter of the crucible, the higher the maximum velocity caused by natural convection.

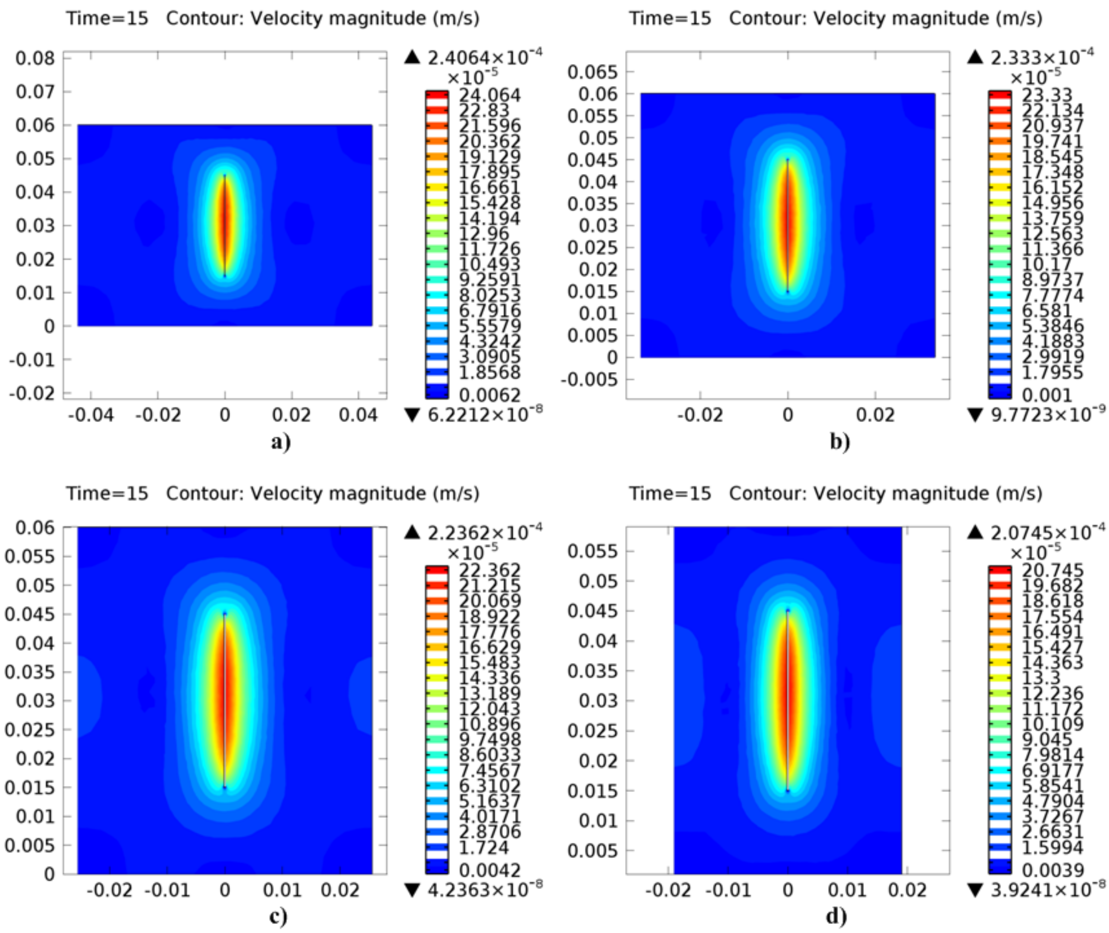


Fig. 6 Velocity Magnitude as a function of crucible diameter a) d = 8.81mm b) d = 6.7mm c) d = 5.1mm d) d = 3.8mm.

Figure 7 shows the velocity field from natural convection after 15 s simulation time. The velocity field is given as a function of the wire position. Figure 7 clearly shows that the velocity is slightly higher (0.016 mm/s) when the wire is out of centre.

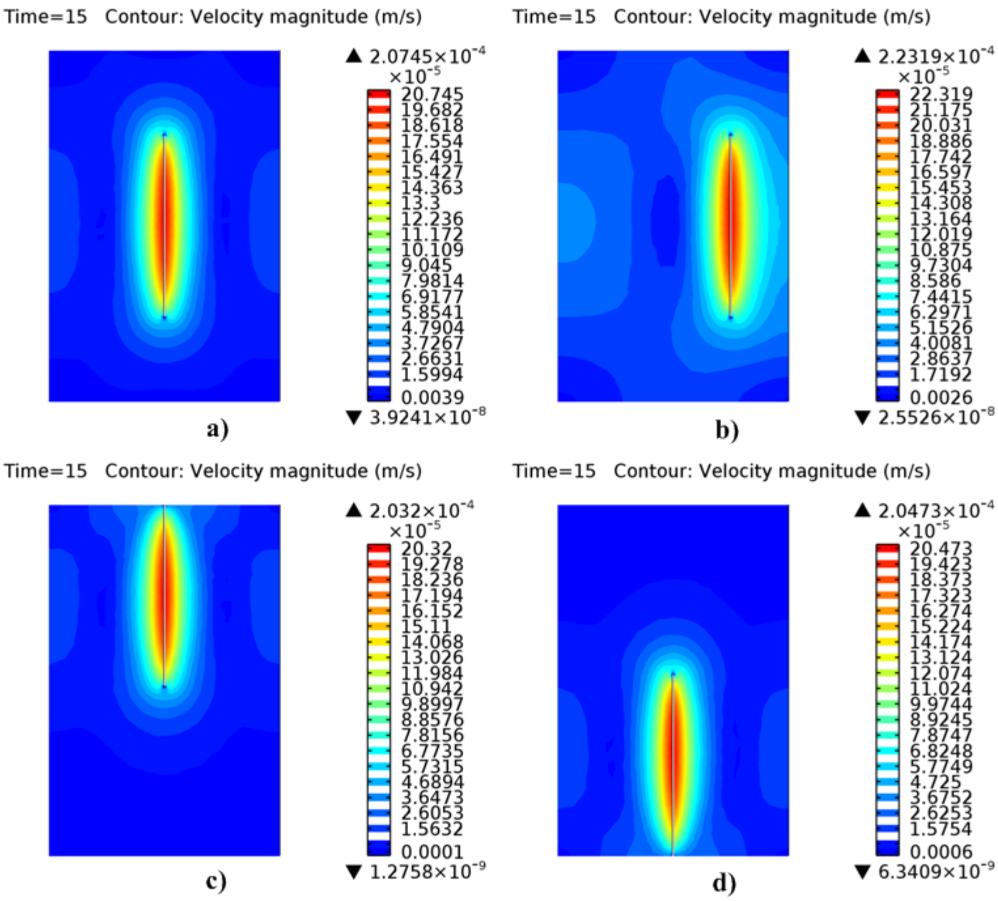


Fig. 7 Velocity magnitude as a function of wire position a) middle b) out of centre c) top d) bottom.

Figure 8 shows the velocity field as a function of the surface cooling on the top of the crucible. The boundary in Figure 8a was set to convective cooling. The heat transfer coefficient used was the built-in COMSOL heat transfer coefficient for air. The reference temperature was set to 298 K. In Figure 8b the boundary on the top of the crucible was set to thermal insulation. As Figure 8a and 8b show, after 15 s simulation time there is no difference in the velocity field. At 303K the cooling on the surface does not seem to have any influence on the velocity field in the liquid.

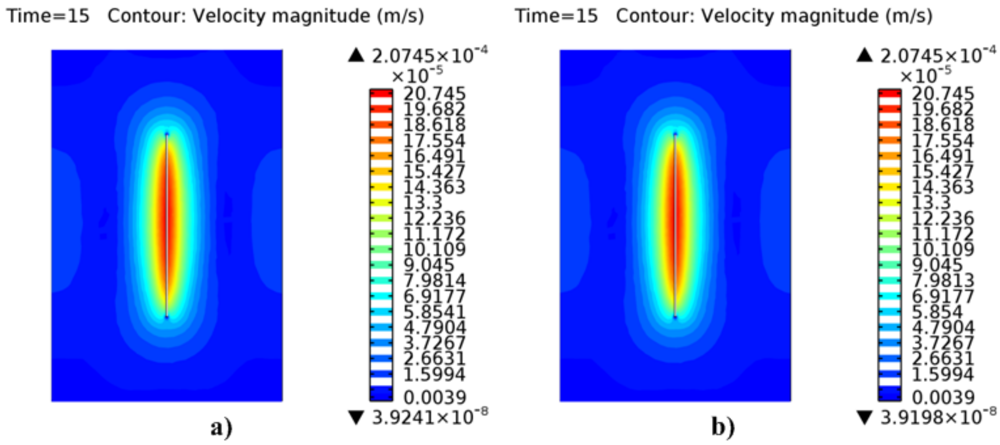


Fig. 8 Velocity magnitude as a function of the top surface cooling a) boundary set to convective cooling b) boundary set to thermal insulation.

Figure 9 shows the velocity field caused by natural convection for the crucible with a diameter of 3.8 mm as a function of time. After 1 second simulation time (Figure 9a) the maximum velocity is 0.018 mm/s and after 300 s simulation time (Figure 9b) the velocity is 1.1 mm/s.

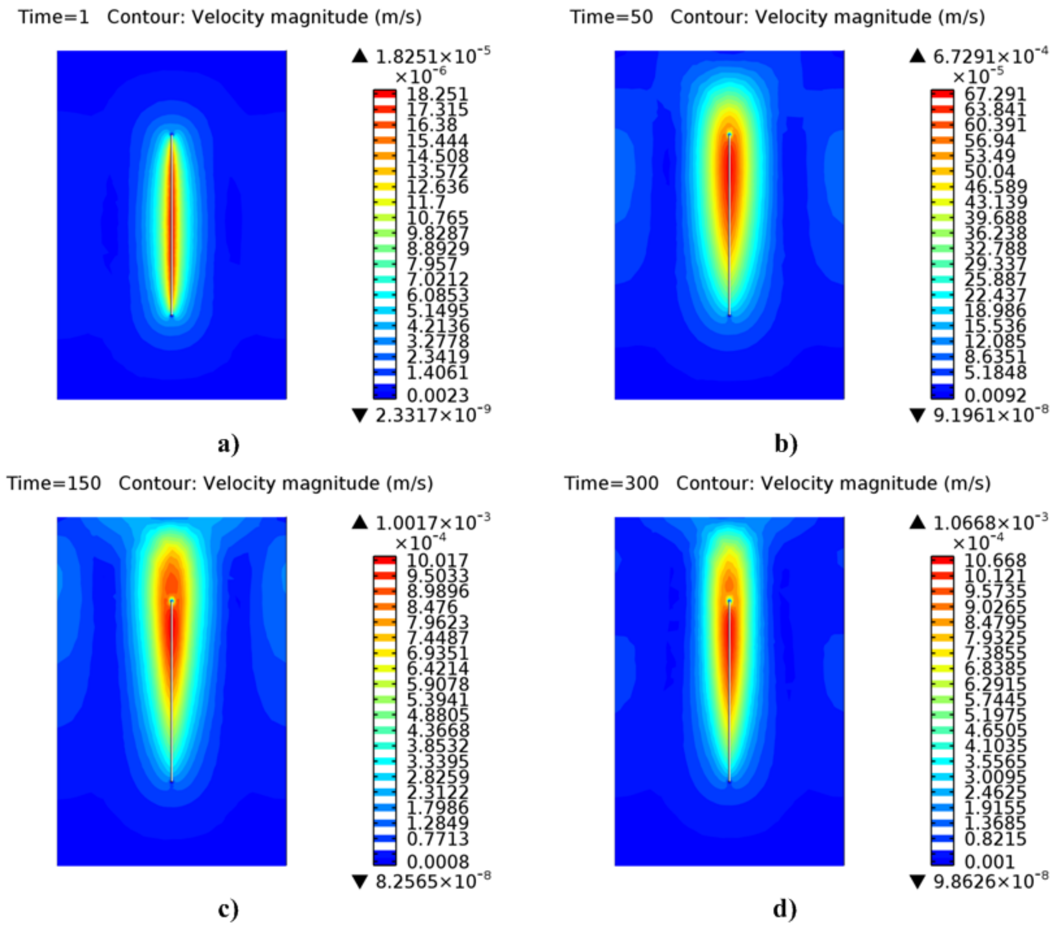
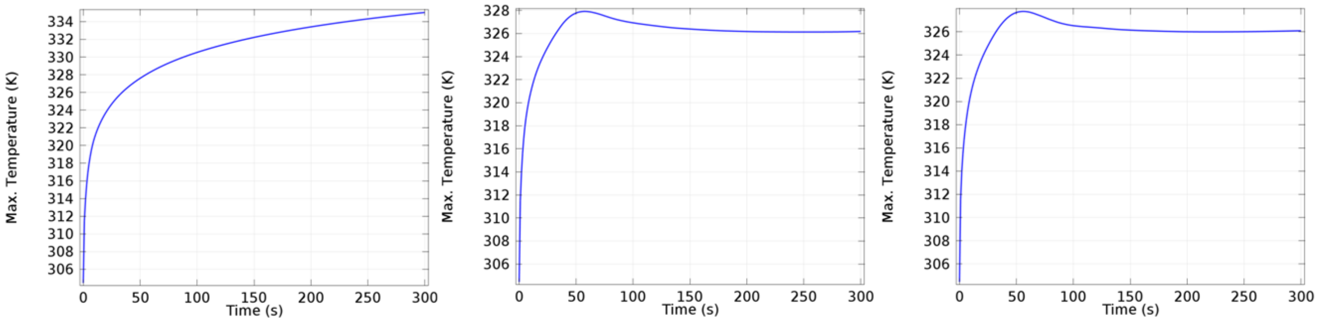


Fig. 9 Velocity magnitude as a function of simulation time a) at 1s b) at 50s c) at 150s d) at 300s.

Figure 10 shows the effect of the natural convection in the crucible on the maximum temperature of the wire as a function of simulation time. Figure 10a gives the maximum temperature profile of the wire without convection effect for 300 s. For this simulation only the conduction was considered and the wire was placed exactly in the middle of the crucible. From the figure it can be seen that the temperature increase of the wire is very high in the beginning. After approximately 10 s the temperature of the wire still increases, but not as fast as at the start. After 300 s simulation time the wire temperature still hasn't reached steady-state status.



a) b) c)

Fig. 10 Effect of natural convection on the maximum temperature of the wire as a function of time a) without convection, wire in the middle of the crucible b) with convection, wire in the middle of the crucible c) with convection, wire out of centre of the crucible

Figure 10b shows the maximum wire temperature with effect of conduction and convection. The wire was placed exactly in the middle of the crucible. Up to a temperature of 328 K the curve is quite similar with the curve simulated without convection. The effect of convection becomes significant after approximately 50 s simulation time. Then the temperature of the wire decreases through the cooling by natural convection. The effect on the temperature of the wire placed off-centre in the crucible is shown in Figure 10c. For this simulation, conduction and convection were taken in consideration. The graph in Figure 10c differs slightly from the one in Figure 10b. The part of the curve in the beginning is almost the same as shown in Figure 10a and 10b. The reached maximum temperature at 50 s simulation time differs slightly from Figure 10b.

6.2 Cold model experimental results

Table 4 lists the thermal conductivity values calculated from measurements for two different wire diameters and four different crucible sizes. All measurements were made at 303 K and for all measurements a constant current of 1.53 A was measured.

Table 4 Calculated thermal conductivity from cold model measurements at 303 K at different diameters of crucible and wire.

Crucible Diameter [mm]	*Measured Source Current [A]*	*Thermal Conductivity d_{wire} = 0.25mm [W/(m*K)]*	*Thermal Conductivity d_{wire} = 0.35mm [W/(m*K)]*
38	1.53	0.24	0.17
38	1.53	0.25	0.15
51	1.53	0.20	0.16
51	1.53	0.26	0.16
67	1.53	0.21	0.17
67	1.53	0.19	0.15
88	1.53	0.20	0.15
88	1.53	0.24	0.18

Table 5 gives the thermal conductivity values calculated from cold model measurements as a function of the position of the wire. All measurements were taken at 303 K and for all measurements a constant current from 1.53 A was measured. For these measurements only the Pt-10%Rh wire with a diameter of 0.35 mm was used.

Table 5 Calculated thermal conductivity from cold model measurements at 303 K at different positions of the wire.

Crucible Diameter [mm]	*Measured Source Current [A]*	*Wire Material*	*Wire Position*	*Thermal Conductivity d_{wire} = 0.35mm [W/(m*K)]*
38	1.53	Pt-10%Rh	top	0.15
38	1.53	Pt-10%Rh	top	0.16
38	1.53	Pt-10%Rh	centre	0.17
38	1.53	Pt-10%Rh	centre	0.15
38	1.53	Pt-10%Rh	bottom	0.14
38	1.53	Pt-10%Rh	bottom	0.17
38	1.53	Pt-10%Rh	off-centre	0.15
38	1.53	Pt-10%Rh	off centre	0.15

Table 6 gives the thermal conductivity values calculated from the cold model measurements as a function of the source current. For all measurements the Pt-10%Rh wire with a diameter of 0.35 mm was used. The measurements were conducted at 303 K. The lowest measured source current was 1.03 A and the highest measured source current was 3.53 A.

Table 6 Calculated thermal conductivity from cold model measurements at 303 K for different source current.

Crucible Diameter [mm]	*Wire Diameter [mm]*	*Wire Material*	*Measured Source Current [A]*	*Thermal Conductivity [W/(m*K)]*
38	0.35	Pt-10%Rh	1.03	0.17
38	0.35	Pt-10%Rh	1.03	0.18
38	0.35	Pt-10%Rh	1.53	0.17
38	0.35	Pt-10%Rh	1.53	0.15
38	0.35	Pt-10%Rh	2.03	0.14
38	0.35	Pt-10%Rh	2.03	0.16
38	0.35	Pt-10%Rh	2.53	0.13
38	0.35	Pt-10%Rh	2.53	0.14
38	0.35	Pt-10%Rh	3.03	0.14
38	0.35	Pt-10%Rh	3.03	0.15
38	0.35	Pt-10%Rh	3.53	0.15
38	0.35	Pt-10%Rh	3.53	0.13

6.3 High temperature experimental results

The calculated thermal conductivity values for the high temperature measurements are given in Table 7. The measurements were conducted at 1773K, 1823K, 1873; and 1923K. The measured slag composition is 40%CaO, 30%Al_2O_3, 18%SiO_2 and 12%MgO given in weight percentage. Two different measurement cycles were realized.

Table 7 Calculated thermal conductivity from high temperature measurements different temperatures.

Slag Composition [Weight %]	*Temperature [K]*	*Thermal Conductivity [W/m*K]*[1]	*Thermal Conductivity [W/m*K]*[2]
40%CaO-30%AL_2O_3-18%SiO_2-12%MgO	1773	0.162	0.179
	1173	0.171	0.178
	1173	0.180	0.177
	1823	0.118	0.111
	1823	0.111	0.110
	1823	0.116	0.116
	1873	0.064	0.061
	1873	0.064	0.060
	1873	0.060	0.063
	1923	0.045	0.046
	1923	0.047	0.035
	1923	0.039	0.033

[1] measured from the highest temperature to the lowest temperature

[2] measured from lowest temperature to the highest temperature

7. Discussion

According to the simulations, the crucible diameter for the high temperature experiments should be as small as possible in order to keep the natural convection as low as possible. The cold model measurements, however, do not show a better result for the smaller crucible diameter in comparison to the larger diameter crucible. The simulation shows that the velocity caused by natural convection at 15 s simulation time (Figure 6) even for the largest simulated crucible diameter (8.81mm) is still very low (0.24 mm/s). For the cold model thermal conductivity calculation only the measured values in the range between 2 till 4 seconds were taken in consideration. This means that the velocity of the natural convection is even lower than calculated in the simulation at 15 s simulation time. Therefore the influence on the thermal conductivity values calculated from measurements is negligible. This is why the cold model measurements show little difference between the different crucible diameters. In order to get closer to the real value during the high temperature conductivity measurement, the crucible diameter was chosen as small as possible.

As seen from Table 4 the thickness of the wire has a much larger impact on the measured and calculated conductivity values. The calculated values from the measurements done with the 0.25 mm wire are approximately 1.5 times higher as the values calculated from the measurements with the 0.35 mm wire. The reason can be due to the follows. The heat generation for the thinner wire per surface area is 2.8 times higher as in the thicker wire, which means that the natural convection starts possibly much earlier than the thicker wire. The values measured with the thinner wire are close to the value found in literature for Rhodosil Silicon Oil 550 (0.14 W/m*K). The discrepancy between the real value and the measured and calculated values could be caused by the age and impurity of the silicon oil or the temperature distribution in the water bath. According to the results from the cold model measurements the thickness of the hot wire for the high temperature thermal conductivity measurements was chosen to be 0.25 mm. The wire thickness most commonly used for thermal conductivity slag measurement in literature [1,2,5] is 0.15 mm. Since the results from our cold modelling show that a slightly thicker wire gives better measurement results the thickness of the wire was chosen as 0.25 mm. On the other hand, if the wire is too thick the heat generation per surface area is not enough to perform reasonable measurements. In some high temperature measurements, a wire with a thickness of 0.5mm was used. Even with increasing current up to 3 A the measured and calculated values did not give reasonable values.

According to the literature [1,2,5] a source current from 1 A to 2 A is commonly used for the high temperature conductivity measurements. Table 6 gives the measured and calculated thermal conductivity values as a function of the source current. The values show the higher the current the closer the thermal conductivity value to the one found in the literature. However, higher current means higher heat generation per surface area and therefore higher risk of convection. Therefore, 1.5 A was chosen as a suitable current for the measurements as a compromise between accuracy and possible higher convection. A systematic and more detailed study might be required to clarify the situation.

The effect of the placement of the wire in the crucible was studied in simulation as well as in the cold model experiments. In Figure 7, it is seen that the velocity and therefore the natural convection are slightly higher when the wire is placed out of centre in the crucible. The placement of the wire close to the top surface or bottom does not have a major effect. To show temperature effect of off-centre placement of the wire the maximum temperature of the wire was plotted over time (Figure 10c) in comparison to the perfect placement in the centre including convection (Figure 10b) and perfect placement in the centre excluding convection (Figure 10a). In all three Figures 10a-10c show that in the beginning the temperature profile looks quite similar. This means if only the measurements in the first seconds are used the effect of the off-centre placed wire on the conductivity values can be neglected. The results of the cold model measurements and calculations given in Table 5 reinforce this conclusion.

8. Summary

The optimal experimental conditions for applying transient hot-wire method to metallurgical slags were studied using the computational fluid dynamics (CFD) method, cold model experiments and high temperature test measurements. On the basis of these studies, a Pt-30%Rh wire with a diameter of 0.25 mm was chosen as a suitable hot-wire for the high temperature thermal conductivity measurements in slag. All other Pt-30%Rh wires had a diameter of 0.5 mm. These wires were sheathed with HfO_2. HfO_2 was found to be a very suitable material for this kind of measurements. It could stay in the slag at high temperatures for a quite long time without losing the function as sheathing material. No dissolution of HfO_2 into slag was observed. On the other hand dense Al_2O_3 failed in the measurements. The diameter of the crucible was found to affect the measurement very little when only the measurements in the first seconds were employed, though smaller crucible theoretically produced better measurements. Also, the slightly higher natural convection caused by the off-centre wire position in the slag could be neglected, if only the measurements in the first seconds were used. The determination of the resistivity and the temperature coefficient of resistance of the wire was found to be a very important procedure to calculate the right value. Even very small changes e.g. in the wire length can cause large differences in the wire resistivity. More studies needs to be performed on the wire diameter and the applied current. Very good reproducibility was obtained in the conductivity measurements of slag using the optimal condition obtained in the present work.

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