Heat transport in the pellet firing process

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Summary

The objective of this project, undertaken from November '98 until June '99 at Hoogovens Research & Development, was to improve the description of the heat transport in a packed bed as found in the pellet firing process and in particular determine the influence of certain process variables, namely temperature, gas flow rate and bed voidage.

Pellets are essentially spherical porous iron ore particles of approx. 12 mm in diameter which are used as charge for the blast furnace process. Pellets are formed by the agglomeration of iron ore grains, a binding substance and some water. After this water has been evaporated, the pellets are fed to the firing process in the form of a packed bed, where they undergo a heat treatment by a flow of gas at a temperature of up to 1600 K. During firing, the individual grains within a pellet sinter, vastly improving its mechanical strength. This in turn largely determines the quality of the pellet for the blast furnace process. The strength of the pellet can be related directly to its temperature history in the firing process. Therefore, a better description of the heat transport in the firing process will result in better control over the pellet production process.

The heat transport in the pellet firing process consist of both heat transport from the gas to the packed bed and heat transport within the individual pellets. Both were investigated in the present work.

The heat transport within a pellet can be described by a heat conduction process, governed by an effective conductivity which was determined experimentally using a tube oven. In the experiments the response of a single pellet was measured to a step change in oven temperature. An effective conductivity was found of 0.3±0.06 W/mK at 700 K which rises to 0.7±0.14 W/mK at 1400 K. This is significantly lower than found in literature. This difference cannot be accounted for by the error and is thought to be caused by a different porous structure of the pellet used in the experiments, possibly due to a lesser degree of sintering. As the accuracy of the estimates was significantly reduced by necessity to include heat transport from the oven to the pellet surface, recommendations are given to further improve the analysis and the experimental method.

The effective heat transport from a flow of hot gas to a packed bed of pellets was investigated by experiments using a pilot scale set up of the pellet induration process. The response of the cylindrical bed to a step change in gas inlet temperature has been measured for a range of temperatures (600-1200K), flows rates (140-700 Nm³/hr) and bed voidages (0.37-0.41). A new approach to measure the bed temperature was used, in which thermocouples were inserted in a pellet. From the experiments, an overall heat transfer could be determined which is in the order of 200 W/m²K and an interfacial heat transfer coefficient was of roughly 500 W/m²K, which lies in the range found in literature. With respect to the dependency of the heat transfer coefficient on temperature, bed voidage and flow rate, no decisive conclusions could be drawn.

The analysis of the experiments has further shown that the overall heat losses in the experiment amount to approximately 70% which was not expected. In order to determine the heat transfer coefficient from the experiments, in which only the temperature on the axis was measured, a hierarchy of models has been developed. In these models, multiple approaches were used to include the effect of the heat losses, but they did not lead to a satisfactory accuracy in the determination of the heat transfer coefficient. It is therefore recommended that the heat losses should be reduced, the radial temperature profile should be measured and if necessary a two dimensional model of the bed should be developed.
Acknowledgements

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M.M.
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1. General introduction

The main objective in this project has been to quantitatively describe the heat transfer to a pellet in the firing section of the pellet production process and its dependency on different process variables. A pellet is in essence a marble-like agglomeration of small iron ore particles of around 1 cm in diameter. In the firing section a packed bed of these pellets is heated by a flow of air at elevated temperatures in order to give them a desired mechanical strength. It has been found that this strength is directly related to the temperature history of the pellet. A better description of the heat transfer is thought to improve the currently used model of the pellet production process. This in turn will lead to better estimates on the temperature history of the pellets and thus to a better control on the strength of the pellets produced.

The following introduction will give a more detailed description of the pellet production process as well as its relation to the iron making process. After this introduction on iron making and pelletising, the objective of this graduation project will be comprehensively explained and how it is related to the actual pellet production.

1.1 The iron making process

In the iron making process, iron ore is effectively converted to iron at elevated temperatures with use of cokes and air, thereby producing a small amount of waste in the form of slag. The process is carried out in a blast furnace as shown in figure 1.1, in which ore and cokes are fed from the top, and hot air or 'wind' is injected near the bottom. Both the produced iron and slag, which are separated by smelting, are drawn from the bottom.

![Blast furnace diagram](image)

Figure 1.1, blast furnace

The global chemical reaction taking place in the blast furnace is the reduction of the iron oxides in the ore by carbon monoxide to form iron and carbon dioxide;

$$\text{Fe}_x\text{O}_y + y\text{CO} \rightarrow x\text{Fe} + y\text{CO}_2$$  \hspace{1cm} (1.1)

The carbon monoxide is obtained from the in-situ gasification of the cokes which is the essential carbon and heating source.
The iron oxides in the ore are dependent on its geophysical (and thus geographical) origin. The two most important forms of iron oxides present in the various commercially available ores are either magnetite \((\text{Fe}_3\text{O}_4)\) or hematite \((\text{Fe}_2\text{O}_3)\). In the iron ore, the oxides are associated with varying portions of waste or gangue material which generally contains silica, alumina and a very small amount (from below 1 to a few percent) of other constituents like compounds of zinc, copper, chromium etc.

The run-of-mine ore, the product of a mining operation, is taken through a number of processes to make it more suitable for blast furnace charge, e.g. crushing, grinding, screening, concentration and blending. In all these processes, including the mining itself an enormous amount of fines (<6 mm) is produced (40-50%) which can not be charged in the blast furnace. The reason for this is that, as can be seen in figure 1.1, air has to be able to flow through the charge, which can be as high as 35m. Particles below 6 mm adversely influence the flow and increase the pressure drop by lowering the voidage of the blast furnace charge.

Since the fines cannot be directly fed to the blast furnace, it is necessary to agglomerate them into lumps which is performed predominantly by sintering and pelletising processes. An important prerequisite of sinter and pellet is their adequate reducibility and strength to withstand handling hazards, and impact, abrasion and compression in the blast furnace as this will result in fines smaller than 6mm which will again decrease the permeability of the blast furnace charge. At Hoogovens both processes are equally important, as the ore charge of the blast furnace is mainly made up of pellets and sinter in varying ratios. This is done in order to have better control over the specification of the charge and thus better and more consistent control of the blast furnace. Furthermore, it improves the flexibility as a constant specification of charge material can be attained by blending ores of different origin. Although both sintering and pelletising are important at Hoogovens, the process of interest in this project is pelletising and the sintering process will not be further looked upon.
1.2 Pelletising process

As explained, pelletising is used to agglomerate the fines. It is especially important for very small fines (<0.05 mm) e.g. from wet concentration, which cannot be processed by sintering but apart from this, pelletising has a number of other advantages as well.

In short, the pelletising process consists of balling up the fines in the presence of moisture and suitable additives into a more or less spherical particle of 11-13 mm in size. These 'green' pellets are subsequently hardened or 'indurated' by a heat treatment 'firing' at temperatures of 1200-1350°C which is called 'firing'. A schematic view of the pelletising process at Hoogovens is given in figure 1.2 and will be discussed below.

For the formation of pellets it is necessary to grind the ore, fed to the plant from the blending fields, to the extent that 60-66% of the material has a size smaller than 0.045 mm. The grinding is done under dry conditions using a bead mill. Other operations in the milling stage include the addition of small amounts of cokes breeze and dolomite for savings on gas use in the induration stage and improvement of metallurgical properties respectively.

Next, the grind ore is transported to the moisturising stage. In the moisturising stage an exact amount of water (9%) and bentonite (0.65%) is added. Bentonite is a clay type substance which has the capacity to take up moisture and has additional adhesive properties. In the following stage, the balling stage, the actual pellets are formed.

The formation is performed in rotating drums (or at the pellet pilot plant: rotating discs with a flange on the rim). The ball formation occurs because of surface tension forces of water and collisions between particles. Initially, small nuclei of pellet are formed on addition of water and the nuclei grow bigger into pellets as they pick up loose grain particles during their travel in the drum. The capillary action of water in the interstices of the grains causes a contracting effect on them. The pressure of water in the pores of the ball is sufficiently high so as to compact the constituent grains into a dense mass. The final stage of the pelletising process is the induration stage where the formed pellets are hardened. It will be looked upon in detail, as the firing process, which is the main objective in this project, is part of this stage.

Figure 1.2, the pelletising process at Hoogovens
1.2.1 The induration stage

The feed of the induration stage, as shown in figure 1.3, are 'green' pellets from the formation stage which are still very fragile as they are held together by the capillary forces of water. They can easily fall apart so before entering the firing section, the pellets are fed over a sieve to separate the broken material outside the size range of 10-15 mm.

At the hart of the induration stage is a moving grate consisting of a closed chain of 187 grate wagons each of which is 1.5 m in length and 3.5 m wide. The pellets are moved on this grate in a packed bed, as shown in figure 1.4, which is approximately 50 cm in height.

The bed can have an average voidage in the range of 0.38-0.41 which is dependent on the operating conditions. The voidage of the bed is influenced by the effectiveness of the sieve before the induration stage as well as by the moisture contents of the pellets which enter the induration section. This variance will be discussed in more detail later, as its influence on the heat transfer will be investigated in this project.

To protect the grate against the high temperatures, the first 5 cm of the packed bed, which is called the hearth layer, consists of indurated pellets. On the grate, which moves with an approximately speed of 4.5 m/min, the pellets are moved through the different sections of the induration stage which has a total length of 123m. In the first two sections, the pellets are dried by hot air, which flows upwards through the bed in the first, and downwards in the second drying section. This is done to prevent re-condensation of moisture in the lower part.
Pelletising process

of the bed. When the pellets are dried but not yet fired, the bentonite helps in maintaining some degree of cohesion among the grains.

The dry pellets are next fed into the firing section which has a length of 52.5m in total. Combined with the speed of the grate, this ensures a residence time of about 12 minutes. In the firing section the packed bed is heated by a flow of hot gas at a temperature of 1320°C and an average flow of 600,000 Normal m³/hr (volume flow calculated at 273K). This results in a superficial velocity in the bed of 0.9 Nm/s, which is about 5 m/s at 1320°C. Within the firing section, subsections can be distinguished, which differ in flow rate of the gas and temperature; in the first subsections the conditions are relatively mild and is called the pre-firing. In the rest of subsections the firing at the highest temperature takes place. The throughput of the firing section is approximately 650 ton/h. After the firing stage, the pellets are cooled by an upward stream of cool air which is subsequently heated and used in the firing zone from the firing zone it is transported to the drying zone. The cooled pellets are sieved for the separation of fines and are ready to be used as charge for the blast furnace.

1.2.1.1 Chemical and physical processes during firing

The most important process within the pellet during firing is the physical/chemical process of sintering of the grains (not to be confused with the sintering production process). During the sintering, the individual grains are bonded. With magnetite, in an excess of oxygen, this bonding is performed by bridging of magnetite grains through hematite crystals formed by the exothermic oxidation of magnetite:

\[ 4\text{Fe}_3\text{O}_4 + \text{O}_2 \rightarrow 6\text{Fe}_2\text{O}_3 \quad (\Delta H = -230 \text{ kJ/mol}) \]  

which will take place at temperatures of 1100-1250 °C. The strengthening of the pellet occurs due to high surface mobility of the atoms during oxidation, re-crystallisation and crystal growth of hematite. When the iron ore contains only hematite, the above mentioned reaction is absent. The strengthening effect is then obtained by a re-crystallisation involving coalescence of ferric oxide particles in the grain. This process needs higher temperatures, usually 1300-1350°C.

As the reaction of magnetite is exothermic, it will supply some of the heat needed to attain the elevated temperatures in the firing section. The cokes breeze, which was added in the milling stage is helpful in this sense as well, as it is burnt and thus supplies extra heat. It also helps to heat up the pellet more uniformly. This is important as the pellet conducts heat badly, which induces a non uniform temperature in the pellet. This in turn leads to non uniform sintering which could invoke so called ‘onion’ formation in the firing section. In onion formation, the outside of the pellet is well sintered, whereas the inside is not, which reduces the mechanical strength under the conditions in the blast furnace of the pellet significantly. As said this will lead to fines and a blast furnace charge that is less permeable to the flow of gas.

1.2.1.2 The 'Branderij Model' (BRAM)

As mentioned, the quality of a pellet is determined, amongst other things, by their strength to withstand handling hazards, and impact, abrasion and compression in the blast furnace. The strength of indurated pellets is related to the cold compression strength and ASTM tumble and abrasion indices.

The essential incentive of the modelling the induration process, as will be discussed below, is that this strength can be predicted from the temperature-time curves of the pellet during the firing process. That is to say, the strength of the pellet in the blast furnace charge can be calculated from the pellet’s temperature history in the induration process.

The control of the induration process is therefore essential in controlling the quality of the pellet. The process control however, is seriously hampered by the absence of on-line measurements of the temperatures in the pellet bed. The temperature in the bed is therefore on-line calculated, using a combination of a flow balance model and a model of the induration process called the 'Branderij Model' (BRAM).
In the flow model, the flows through the different stages of the induration process are calculated from measurements on the process fans. These calculated flows are then used in the induration model, in which the temperature of the pellets is calculated on-line as a function of the section and depth in the bed. The stage dependency is achieved by modelling a section of the bed while it is going through the different stages of the induration process subsequently. The dependency of the depth in the bed is achieved by dividing this section of the bed vertically into several layers. The heat transfer from the gas to the pellet $\Phi_{\text{gas-pellet}}$ is modelled by an overall heat transfer coefficient $h_{\text{tot}}$ and the difference between the average temperature of the gas $\langle T \rangle_{\text{gas}}$ and the average temperature of the pellet $\langle T \rangle_{\text{pellet}}$ in each layer as

$$\Phi_{\text{gas-pellet}} \propto h_{\text{tot}} \cdot (\langle T \rangle_{\text{gas}} - \langle T \rangle_{\text{pellet}})$$

(1.3)

This heat transfer coefficient combines the heat transport both to the pellet and within the pellet and is assumed constant in each of the sections of the induration process.
1.3 Objective of the project

In general, the objective of the project is to improve the way in which the heat transport from gas to pellet in the firing section is modelled in BRAM by investigating the dependency of this overall heat transfer coefficient on the process conditions in the firing section. Although the focus has been on the firing section, similar conditions are found in the drying and cooling zone, so findings on the heat transport are also likely to be used in the modelling of those sections. Improving the model could lead to better control of the induration stage and therefore to a better control of the pellet quality. The investigations are part of long term project on the improvement of pellet quality at Hoogovens in co-operation with three other European companies within the European Community of Coal and Steel.

The heat transfer coefficient is important as variations in its value are expected to lead to different temperature profiles within the bed as well as within the pellet, especially in deeper regions of the bed. As in the firing stage the pellets are heated by a downwards gas flow only, the quality of the pellets at the bottom of will determine the overall quality of the pellets leaving the firing section. The heat transfer coefficient could therefore significantly influence the predicted quality of the induration stage.

The process conditions which are most likely to vary within the firing section are the temperature and the flow. For the temperature, this variation is obvious as in the firing section, the pellet bed temperature is increased from about 100°C at which the drying takes place up to the maximum temperature of 1320°C. These temperature differences will also induce flow differences within the bed. There is also significant variation in the bed voidage reaching the firing section although once within the firing section, it does not notably change. This has two reasons, first of all the voidage is lowered due to fines. Fines are introduced in the induration stage either by insufficient sieving after the balling stage (the effectiveness of the sieve is dependent on the throughput) or by breakage of pellets within the induration stage. The bed voidage is also influenced by the variation in the moisture content of the pellet; more moisture lessens the capillary forces and makes the pellet more able to deform and thus for the bed to be compacted.

It was mentioned that the overall heat transfer coefficient covers both transport to the packed bed and within the pellet. In the investigation these two modes of transport will be looked upon separately in chapter 2 and chapter 3 respectively. The reason for this is two fold; first of all, some mechanisms are primarily dependent on the surface temperature of the pellet and with the pellet being a bad conductor of heat, this could be significantly different from the average pellet temperature. Secondly, specific quality issues in the pellet are dependent on the internal temperature profile within pellet, e.g. onion formation. It is therefore, important to be able to predict the internal heat transport.

The investigations were carried out at Hoogovens, where an electric tube oven has been available for experiments on individual pellets. For experiments on a packed bed of pellets under the conditions of the firing stage, a pilot scale set-up has been used.
1.4 References

2. Internal pellet heat transport

2.1 Introduction and objective

The heat transport within a pellet can be described effectively as a heat conduction process, governed by the effective conductivity ($\lambda$), the density ($\rho$) and the specific heat ($c_p$) of the pellet. Of these properties, the density and specific heat are easily measured and well known\(^1\). The objective of the investigations will be to determine the effective conductivity of the pellet. On the subject of pellet conductivity, several authors have published values and temperature dependencies which differ significantly according to figure 2.1 and 2.2.

![Figure 2.1, conductivity temperature dependency found by Beer\(^3\),
(conversion factor to W/m-K: $\times 4.18$)](image1)

![Figure 2.2, conductivity temperature dependency found by Bratchikov\(^5\),
(conversion factor to W/m-K: $\times 1.16$)](image2)

This difference could well be caused by the fact that a pellet does not have a homogenous but a micro-porous structure. The pores give rise to mechanisms of heat transport other than solid conductivity such as gas conductivity and radiation in the pores. These mechanisms are not only influenced by the total volume of pores but by their nature (e.g. tortuosity and connectivity) and size distribution as well. The form in which the pellet material is used in the experiment, will thus influence the effective conductivity found.

In the research of Beer\(^4\) and Bratchikov\(^5\) a body composed of crushed pellets was used. In order to correctly predict the conductive properties of a pellet, the test material should be in the shape and state of a pellet. Therefore, using a single pellet, experiments have been conducted to determine the dependency of the conductivity on temperature.
The effective conductivity can be determined by analysing the transient behaviour of a pellet to a step-change in pellet surface temperature. In the apparatus available for the experiments, described in the experimental section, it was only possible to induce a step-change in the temperature of the environment of the pellet. It was therefore necessary to investigate in detail, the heat transport from the heating source to the pellet surface. This investigation is found the theory section. The findings are used in the analysis of which the results and conclusions are given in the subsequent sections.
2.2 Experimental

2.2.1 Experimental set-up
For the experiments on the effective conductivity of a pellet, an electrically heated tube oven was available, as shown in figure 2.3 and documented in table 2.1. The oven operated at constant temperature of 1070°C. However, using the temperature profile present along the length of the oven tube, experiments could be conducted at different temperatures of the surroundings. The temperature profile which was measured with an unprotected thermocouple is given in figure 2.4.

![Figure 2.3, tube oven](image)

![Figure 2.4, temperature profile in the tube oven](image)

Table 2.1, Tube oven properties

<table>
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<tr>
<th>Property</th>
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<td>Temperature range (°C)</td>
<td>20-1100</td>
</tr>
<tr>
<td>Tube material</td>
<td>Al₂O₃ (99.7%)</td>
</tr>
<tr>
<td>Inner diameter (mm)</td>
<td>23</td>
</tr>
<tr>
<td>Outer diameter (mm)</td>
<td>30</td>
</tr>
<tr>
<td>Tube length (mm)</td>
<td>1400</td>
</tr>
<tr>
<td>Tube emissivity</td>
<td>0.9-0.5</td>
</tr>
</tbody>
</table>

It was attempted to induce a specified step-change in the surface temperature of a pellet by a swift change in its axial position within the oven. It was further assumed that with a small change in temperature of 50K, pellet properties could be considered independent of temperature. By measuring the response of a pellet to a step-change of approximately 50K, at different temperatures, the temperature dependency of the conductivity was determined. The change in position necessary to obtain the required temperature change, was approximated using the profile in figure 2.4. This profile has not been used in further analysis of the experiments.

In the experiments, approximately spherical pellets were used as specified in table 2.2. The pellets consisted mainly of hematite (Fe₂O₃), in order to prevent the occurrence of chemical reactions.

![Internal pellet heat transport](image)

Table 2.2, Pellet properties

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material</td>
<td>Brasil ore: Fe₂O₃ (99.7%)</td>
</tr>
<tr>
<td>Outer diameter (mm)</td>
<td>12.2 ± 0.1</td>
</tr>
<tr>
<td>Density (kg/m³)</td>
<td>3280 ± 100</td>
</tr>
<tr>
<td>Porosity</td>
<td>0.35±0.05</td>
</tr>
<tr>
<td>Pore size (μm)</td>
<td>0.4 / 4.7</td>
</tr>
<tr>
<td>Specific heat (J/kg-K)</td>
<td>800-1000</td>
</tr>
<tr>
<td>Emissivity</td>
<td>0.82-0.85</td>
</tr>
</tbody>
</table>
Experimental

To facilitate the insertion of, and improve the contact with a thermocouple, the pellet was still 'green' (soft and moist) before the experiments. The actual experiments were conducted, only after the pellets had been dried and indurated, to prevent heat effects in the form of evaporation and sintering.

2.2.2 Measurement method

As mentioned, it was necessary in the experiments to shift a pellet to different positions along the length of the oven and measure its temperature response. To achieve this, a probe was constructed as shown in figure 2.5. It could be inserted in the oven tube and slid backwards and forwards in the oven to bring a pellet to the desired position. The dimensions of the probe are such that the pellet is positioned on the axis of the tube.

Figure 2.5, probe used in the experiments

The response of the pellet in each of the experiments was measured, using a small thermocouple put into the centre of the pellet; a technique successfully used in other research on pellets ⁶. The temperature of the thermocouple was logged with a frequency of 1 Hz, using an isolated measuring pod (imp) connected to a personal computer. The imp measured a signal of ±12V and used a 12 bits AD converter. Its specified sensitivity is <0.1°C for the type of thermocouple used ¹⁰. Further properties of the thermocouple are supplied in table 2.3 and a cross section is given in figure 2.6.

Table 2.3, Thermocouple properties ¹¹,¹²

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
<td>K-type (NiCr-Ni)</td>
</tr>
<tr>
<td>Outer diameter (mm)</td>
<td>1</td>
</tr>
<tr>
<td>Length (mm)</td>
<td>1000</td>
</tr>
<tr>
<td>Sheath material</td>
<td>Stainless steel (310)</td>
</tr>
<tr>
<td>Temperature range (°C)</td>
<td>277-1260</td>
</tr>
<tr>
<td>Limits of error (based on °C)</td>
<td>±0.4%</td>
</tr>
</tbody>
</table>

Figure 2.6, cross section of a thermocouple ¹³
An anticipated problem with this method is a net conductive heat flow into the pellet through the thermocouple, as a thermocouple is especially sensitive to temperature gradients close to its tip\textsuperscript{13}. Insulation of the thermocouple outside of the pellet by ceramic tubes, specified in table 2.4 and figure 2.5, was used to prevent this effect. Finally, an extra thermocouple positioned within the insulation (included in figure 2.5) was used to monitor the effect.

### Table 2.4, Insulation tube properties \textsuperscript{14}

<table>
<thead>
<tr>
<th>material</th>
<th>(\text{Al}_2\text{O}_3) (99.7%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>outer diameter (mm)</td>
<td>2.7 / 7.4</td>
</tr>
<tr>
<td>inner diameter (mm)</td>
<td>2.0 / 2.7</td>
</tr>
<tr>
<td>conductivity (W/m-K at 1000°C)</td>
<td>4.5</td>
</tr>
<tr>
<td>specific heat (J/kg-K)</td>
<td>840-1170</td>
</tr>
</tbody>
</table>

It is noted that use of the probe and especially the ceramic insulation has an undesired side effect on the measurement, as it will partly shield one side of the pellet from radiation. However, no alternative means of insulation was available and the effect of shielding is considered less important than the heat losses. Furthermore, it is likely that the shielding of the pellet will be compensated for, by conduction and radiation from the insulation and thermocouple.
2.3 Theory

2.3.1 Effective conductivity

As a pellet is a micro-porous structure, different mechanisms are available for internal heat transport, each with their own magnitude and temperature dependency. First of all, heat can be transported by conduction within the solid phase. A second and a third mechanism are solid conduction in combination with either convection or radiation within a pore. Finally it is possible that heat is transported by convection within the pores. The different mechanisms are summarised in figure 2.7.

In a description of heat transport within a pellet, the different mechanisms can be lumped into one parameter: the effective conductivity of the pellet ($\lambda_{\text{eff}}$). The internal heat transport can than be further regarded as conduction through a single homogenous phase governed by this effective conductivity. Within a pellet, the different mechanisms are expected to operate parallel to each other but depending on the conditions, certain mechanisms will prevail over others.

In general, the pores and temperature differences in the pellet are too small to induce convection. This effect will only become important in pores with a diameter over 5mm. As a result, convective heat transport within the pellet will be neglected.

Heat transport by radiation within the pores is achieved by electromagnetic waves and is highly dependent on temperature. Furthermore, it is dependent on the pore size, the structure of the pores, the emissivity of the substance and the structure of the radiating surfaces. As gasses of one or two atoms hardly influence the radiation, gas-filled pores will pose a relatively negligible resistance to heat transport at higher temperatures compared to conduction through the pores. At which temperature radiation becomes important can be estimated from a simple analysis given in figure 2.8.

In this situation the heat flow by conduction through the gas and radiation can be approximated by

$$\phi_{\text{conduction}} = A\left(\frac{\lambda_{\text{pore}}}{d_{\text{pore}}}\right)\Delta T$$  \hspace{1cm} (2.1)

$$\phi_{\text{radiation}} = A4\sigma T^4\Delta T$$  \hspace{1cm} (2.2)
respectively, in which $A$ is the area of the solid surfaces, $d_{\text{pore}}$ the median pore diameter ($\approx 5\mu m$, see table 2.2), $\Delta T$ the temperature difference over the pore, $\sigma$ is the Stefan-Boltzman constant ($5.67 \times 10^{-8} \text{ W/m}^2\text{K}^4$) and $\lambda_{\text{pore}}$ the conductivity of the gas in the pore which is assumed to be air. In the derivation of the radiation both solid surfaces are taken as black bodies.

From equation 2.1 and 2.2 it can be derived that radiation becomes more important than conduction at

$$T > \left( \frac{\lambda_{\text{pore}}}{d_{\text{pore}}^4 \sigma} \right)^{1/3}$$

(2.3)

2.3.1.1 Estimation of the effective conductivity

With the conductivity of the gas pore given in figure 2.9 and an estimate of the pore size from table 2.2 it is calculated that radiation is more important than gas conduction at temperatures above approximately 3000 K. It can be concluded that radiation is not an important mechanism of heat transport within the pellet, radiation in the temperature range looked upon. It is therefore omitted from further analysis.

For the situation of a pellet, this leaves only conduction within the solid either or not in combination with conduction through the gas in the pores. Assuming only transport of heat through conduction in the pore and the solid, the expected range of the effective conductivity can be estimated by considering two extreme cases as shown in figure 2.10.
In this analysis, the porosity of the pellet is assumed 30%. The conductivity of the solid phase \( \lambda_{\text{solid}} \) (hematite) is estimated around 2 W/mK\(^1\) and the conductivity of the pore can be found in figure 2.9. In the first case, conduction through the pore and the solid within a pellet operate completely parallel. This will result with the aforementioned values of the conductivity and pellet porosity to a value for the effective conductivity of approximately 1.2 W/mK at 400K. In the second case, conduction through the air and the ore within a pellet operated completely in series. With the same assumption as in the former case, this leads to an effective conductivity of approximately 0.12 W/mK. Both Bratchikov and Beer predict an conductivity of approximately 0.6 W/mK at 400 K which is consistent with these estimations.

From this low value of the conductivity, it can be concluded that not all the heat is transported through the solid phase only parallel to the pores but that the pores can be considered more in series with the solid conduction. The most likely reason for this lies in the 3D structure of the pellet. The pellet can be seen as an agglomeration of small (spherical) iron ore particles (<45 nm). During firing, these particles are connected to each other through sintering. When after sintering the individual have only a small portion of there area connected to surrounding particles, this leads to a bottle-neck in the conductive heat transport through the solid. In the areas where the particles are not connected but very close to each other, the conduction through the gas can become competitive with the solid conductivity. It can be concluded that the conductivity in the pellet is dependent on the 3D structure of the solid and the pores which is difficult to predict.\(^1\)

2.3.1.2 Temperature dependency

Conduction through the gas is achieved by collision with other gas molecules. The conductivity associated with this mechanism, is roughly the same for most gasses and very low in comparison with solid conductivity. The conductivity of gas increases with increasing temperatures \( (\propto T) \). In inorganic, non-porous substances internal heat transport is dominated by conduction through phonons which are elastic vibrations of the lattice moving through the solid in waves. In general, phonon conductivity decreases with increasing temperature \( (\propto 1/T) \).

The temperature dependency of the effective conductivity will be a superposition of the dependencies of the different mechanisms. Using the model of figure 2.7 from which radiation and convection in the pores are omitted, it can be derived that the temperature dependency would be of the form

\[
\lambda_{\text{eff}} = \frac{\alpha}{T} + \frac{1}{\left( T + \frac{1}{\beta} \right)^\gamma T}
\]

\[(2.4)\]
in which it was assumed that the resistances to heat transfer are inversely proportional to the conductivity. Parameter $\alpha$ is related to the area and path length of the conduction through the solid alone, $\beta$ to the area and path length of conduction through the solid in series with conduction through the pores for which the $\gamma$ relates to the conductivity of the gas in the pores and the path length.

This derivation is only valid when these parameters and thus the path length and area for conduction are independent of temperature. In fact, due to the 3D structure mentioned above, this assumption does not have to be valid. This is reflected in the difference in results of Bratchikov and Beer, as shown in figure 2.1-2.2. Both also show an initial conductivity of around 0.6 W/mK. However, the influence of temperature is totally different. Whereas in the experiments of Beer the conductivity decreases slowly with increasing temperature, it rises significantly in the experiments of Bratchikov over the same temperature range (400-900K). This could be the influence difference in internal structure of the pellet material used. This is supported by the fact that in both cases no satisfactory fit could be obtained using equation 2.4. However, equation 2.4 could give insight into the possible mechanisms at work in both experiments. Apparently, the conductivity in the experiments by Bratchikov is limited by the conduction of the gas in the pores, whereas in case of the experiments of Beer, the solid conductivity is dominant. An explanation could be that the pellet material in the experiments of Beer was much better sintered. In that case, the conduction through the solid only would be dominant. The results of Bratchikov can be explained in turn with a badly sintered sample in which the pores between the individual solid particles dominates.

It can be concluded from the analysis of the effective conductivity that it can not be predicted beforehand what the temperature dependency of the conductivity of the pellets used in this project will be and has be determined experimentally.

### 2.3.2 Resistance analysis

The aim in the experiments was to induce a step-change in pellet surface temperature. In practice, only the temperature of the environment can be changed in a step-wise fashion. It has to be investigated first, how this influences the intended step-change on the pellet surface.

This is done by comparing the importance of heat transport from the oven wall to the pellet surface (external), to the importance of heat transport within the pellet (internal). The possible mechanisms of external heat transport are radiation, forced and free convection and conduction through the air, whereas the mechanism of internal transport is treated in the previous section.

\[ \Phi_q = \frac{1}{R_{\text{total}}} \Delta T \]

\[ 1/R_{\text{external}} = \sum 1/R_i \]

\[ R_{\text{total}} = R_{\text{external}} + R_{\text{internal}} \]

Figure 2.11, resistances to heat transport in the experiment, where $R$ denotes resistance (K/W)
The importance of the different mechanisms is evaluated by comparing their resistance \((1/h)\) following from Newton's law of cooling. The external resistances can be thought of as parallel to each other, in series with the internal resistance of the pellet (see figure 2.11). All resistances are evaluated per unit area of pellet surface, in which the pellet is assumed to be spherical.

2.3.2.1 *Internal resistance*
An expression approximating the internal resistance of a pellet can be derived from the equation describing the heating of a sphere with constant surface temperature, by assuming that the process has continued for sufficiently long time\(^{19}\). When it is further assumed that the solid properties are independent of temperature, for the internal resistance \(R_{\text{internal}}\) is found,

\[
R_{\text{internal}} = \frac{1}{h_{\text{internal}}} = \frac{3r_{\text{pellet}}}{\pi^2 \lambda_{\text{pellet}}} \tag{2.5}
\]

where \(h_{\text{internal}}\) denotes the internal heat transfer coefficient, \(r_{\text{pellet}}\) the pellet diameter and \(\lambda_{\text{pellet}}\) the effective pellet conductivity.

2.3.2.2 *External conduction resistance*
In the derivation of an expression for the external resistance \(R_{\text{conduction}}\) through conduction of the gas it is assumed that the geometry of a pellet in a tube can be approximated by a geometry of two concentric spheres of uniform temperature. When furthermore, a steady temperature profile between the pellet and the oven is assumed the following relation holds,

\[
R_{\text{conduction}} = \frac{1}{h_{\text{conduction}}} = \frac{(r_{\text{oven}} - r_{\text{pellet}}) r_{\text{pellet}}}{\lambda_{\text{air}} r_{\text{oven}}} \tag{2.6}
\]

where \(h_{\text{conduction}}\) denotes the external conduction heat transfer coefficient, \(r_{\text{oven}}\) the diameter of the oven and \(\lambda_{\text{air}}\) the conductivity of the air. The relation of equation 2.6 is derived in appendix A.

2.3.2.3 *External radiation resistance*
In the expression of the external resistance through radiation \(R_{\text{radiation}}\), the pellet in the tube is modelled as two concentric, grey and diffuse radiating spheres of uniform temperature\(^{20}\). Finally the fourth-order temperature term is linearised to express the relation in the form of Newton’s law of cooling which leads to,

\[
R_{\text{radiation}} = \frac{1}{h_{\text{radiation}}} = \frac{1}{4\sigma T_{\text{avg}}^3} \left[ \varepsilon_{\text{pellet}} + r_{\text{pellet}}^2 \varepsilon_{\text{oven}} - (1 - \varepsilon_{\text{pellet}}) (1 - \varepsilon_{\text{oven}}) \right] \tag{2.7}
\]

wherein \(h_{\text{radiation}}\) denotes the external radiation heat transfer coefficient, \(\varepsilon_{\text{oven}}\) the emissivity of the oven tube surface, \(\varepsilon_{\text{pellet}}\) the emissivity of the pellet surface, \(\sigma\) the Stefan-Boltzman constant and \(T_{\text{avg}}\) the average of oven en pellet surface temperature.

2.3.2.4 *External convection resistance*
Forced convection can be considered negligible, as the air flow through the pipe is absent. Its resistance is therefore very large compared to other mechanisms.

The resistance for heat transfer through free convection \(R_{\text{free conv}}\) is estimated using relations for parallel flat vertical plates\(^{21}\).
Theory

\[ R_{\text{free conv}} = \frac{1}{h_{\text{free conv}}} = \frac{Nu_{\text{free conv}} \lambda_{\text{air}}}{L_{\text{plate}}} \]  
(2.8)

\[ Nu_{\text{free conv}} = 0.52 \cdot (Gr \cdot Pr)^{0.25} \]  
(2.9)

wherein \( h_{\text{free conv}} \) denotes the external free convection heat transfer coefficient, \( L_{\text{plate}} \) the vertical height of the plate. Also the Prandtl number in air (Pr) and the Grashof number (Gr) are introduced. In the calculation of the resistance, the geometry of a pellet in the tube is modelled as two pairs of parallel vertical plates, whose length equal the diameter of the pellet. The two plates are separated by a distance equal to the distance between the pellet surface and the oven tube surface.

Over the entire range of experimental temperatures, the resistance of free convection is much higher than the resistance of conduction and radiation as can be seen in figure 2.12. The complete analysis is given in appendix B. It is noted that in principle the equation 2.6 for natural convection does not hold for the experimental conditions, as the Grashof number is too small. This indicates that the driving force of natural convection is very small. Therefore, it can be assumed that the resistance of natural convection is even higher than given in figure 2.12, as predicted by equation 2.6-2.7. Accordingly, the effect of natural convection will be neglected in further analyses.

As to resistance of conduction and radiation, it can be concluded that the transport through radiation is dominant although conduction can not be neglected, especially at lower temperatures.

![Figure 2.12, external resistances](image)

The conduction and radiation resistance are parallel and can be replaced by a total external resistance. The internal resistance of the pellet is very sensitive to the pellet conductivity. In figure 2.13 both resistances are compared for the approximate extreme values in the expected range of the internal conductivity from figure 2.1 and 2.2. At low temperatures the external resistance is dominant but for higher temperatures the internal resistance is dominant for the expected range of the effective conductivity. The temperature at which this change occurs is higher for higher conductivities.
2.3.3 Modelling the experiment

As both external and internal resistance are of importance, the heat transport in both sections has to be included in a model of the experiment. However, to evaluate the influence of these boundary conditions on the results of the model, analyses have also been conducted using simpler boundary conditions. The first simple analysis assumes constant temperature of the pellet surface and the second external transport by radiation exclusively. In a third analysis, the transport of conduction is introduced next to the transport by radiation.

2.3.3.1 Internal heat transport

Conduction through a single, homogenous phase, of spherical shape, can mathematically be represented by the following differential equation,

\[
\left( \frac{\lambda}{\rho c_p} \right)_{\text{pellet}} \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right)
\]  

(2.10)

wherein \( r \) denotes the radial position within the pellet, \( t \) the time, \( \rho \) the density and \( c_p \) specific heat. For the conductivity \( (\lambda) \), the effective conductivity is used. These pellet properties are assumed to be independent of temperature during an experiment.

One of the boundary conditions of this equation is, that due to symmetry of the problem, there will be no heat flux through the centre of the pellet. The other boundary condition, at the outer surface of the pellet, will have to be supplied by the external transport processes.

2.3.3.2 Constant surface temperature boundary condition

When the external resistance is negligible compared to the internal resistance, the external transport can be considered infinite, resulting in the boundary condition of a constant surface temperature. For a step-change in the surface temperature, an exact solution of equation 2.10 for the temperature at a given position or for the volume average temperature can be derived as \(^9\)

\[
\Theta(r) = \left( \frac{T_{\text{pellet}}(r) - T_{\text{pellet}} \mid r=r_{\text{pellet}}}{T_0 - T_{\text{pellet}} \mid r=r_{\text{pellet}}} \right) = \sum_{i=1}^{\infty} C_i(m_j) \cdot f_i(m_j, r) \cdot e^{-\frac{m_j^2}{(\rho c_p)_{\text{pellet}} r^2} \cdot t}
\]  

(2.11)

---

### Figure 2.13

Total external resistance compared to internal resistance for an effective pellet conductivity of 0.1 and 1 W/m-K

---

**Internal pellet heat transport**

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\[
\bar{\Theta} = \left( \frac{T_{\text{pellet}}(r \to \text{pellet}) - T_{\text{pellet}}(r \to \text{pellet})}{T_0 - T_{\text{pellet}}(r \to \text{pellet})} \right) = \sum_{i=1}^{\infty} C_i(m_i) \cdot D_i(m_i) \cdot e^{-\frac{m_i^2 \lambda_{\text{pellet}}}{\rho_{\text{pellet}} c_{\text{pellet}} r_{\text{pellet}}^2} t}
\]  
(2.12)

in which \(T_0\) denotes the initial temperature of the pellet, \(\Theta\) and \(\bar{\Theta}\) the dimensionless pellet temperature and average dimensionless pellet temperature respectively. The terms in these equations can be calculated using the following relations

\[
m_i = \tan(m_i)
\]  
(2.13)

\[
C_i(m_i) = 2 \frac{\sin(m_i) - m_i \cos(m_i)}{m_i - \sin(m_i) \cos(m_i)}
\]  
(2.14)

\[
D_i(m_i) = 3 \frac{\sin(m_i) - m_i \cos(m_i)}{m_i^3}
\]  
(2.15)

\[
f_i(m_i, r) = \frac{\sin \left( m_i \frac{r}{r_{\text{pellet}}} \right)}{m_i \frac{r}{r_{\text{pellet}}}}
\]  
(2.16)

It can be shown with figure 2.14 that for sufficient long times, both equation 2.11 and 2.13 obey Newton’s law of cooling

\[
\bar{\Theta} = \Theta(r) = e^{-\frac{A}{\rho c_P r_{\text{pellet}} \lambda_{\text{pellet}}} \frac{t}{\tau}}
\]  
(2.17)

in which \(V\) denotes the pellet volume and \(A\) the pellet surface area and \(\tau\) the characteristic time of the heating process.

Figure 2.14, exact solution of the heating of a sphere with constant surface temperature

For a spherical particle the relation between the internal heat transfer coefficient and the conductivity is given by equation 2.5. From an experiment with a step-change in the temperature of the surroundings, the conductivity can be found from the slope of the logarithmic dimensionless temperature response curve. For the expected range of effective conductivities, the characteristic response time is expected to be 10-100 seconds.
2.3.3.3 External heat transport

As was concluded from the analysis of external resistance, the external heat transport consists of both conduction of the gas and radiation of the oven wall. These two mechanisms operate parallel to each other, so the sum of their respective heat flows will be the boundary condition needed in the equation for the internal process. In a simplified analysis, only the radiation boundary condition can be used.

As to the radiation heat flow from oven wall to the pellet, the geometry of the experiment is modelled in the same way as in the resistance analysis; as two concentric spheres (compare with equation 2.5). The temperature dependency however, does not have to be linearised resulting in,

\[ \dot{q}_{\text{radiation}} = A_{\text{pellet}} \frac{\sigma}{1 + \left( \frac{1 - \varepsilon_{\text{pellet}}}{\varepsilon_{\text{oven}}} \right)} \left( T_{\text{oven}}^4 - T_{\text{pellet}}^4 \right) \left( r_{\text{oven}} - r_{\text{pellet}} \right) \]  

(2.18)

The heat flow to the pellet by conduction through the air is again modelled by assuming the concentric sphere geometry. Furthermore, it is assumed that the temperature profile in the air between the pellet and the oven wall is fully developed at all times.

\[ \dot{q}_{\text{conduction}} = \frac{A_{\text{air}} a_{\text{oven}}}{r_{\text{oven}} - r_{\text{pellet}}} \left( T_{\text{oven}} - T_{\text{pellet}} \right) \]  

(2.19)

The assumption of a fully developed temperature profile can be assessed by comparing the times needed for the development of the temperature profile in the air and within the pellet. A criteria for assessment of this time is by looking at the Fourier number of the process \( (Fo=0.1) \). Using this criteria it will take approximately 0.05 and 100 seconds for the full development of a temperature profile in the air and pellet respectively, which renders the assumption of a fully developed temperature profile correct. This is illustrated in figure 2.15 and calculated in appendix C.

![Figure 2.15, comparison of heat penetration time in gas and pellet for the expected range of the effective conductivity](image)

Internal pellet heat transport

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2.4 Numerical implementation

As no simple exact solution is available for equation 2.10 with either, or both radiation and conduction boundary conditions, it has to be solved numerically. The model equation is a parabolic partial differential equation (pde), first order in time (t) and second order in the space co-ordinate (r). The method of solving this kind of equation is in two steps. First, the pde is made discrete in the space co-ordinate, before the resultant ordinary differential equation (ode) is numerically integrated over the required time span.

2.4.1 Discretisation method

The first step in the discretisation of a pde is the construction of a grid which consists of a finite number of points at which the value of the dependent variable, in this case the pellet temperature, will be determined.

Different discretisation methods exist but in this project the 'control volume method' is used as it is easy to understand and lends itself to direct physical interpretation. In this method the calculation domain is divided into a number of non-overlapping control volumes, such that there is one control volume surrounding each grid point. For the geometry of a sphere, combined with a one dimensional pde, these control volumes have the shape of concentric, spherical slabs. In this project the grid points are located in the middle of the control volume. The pde can now be regarded as originating from a thermal energy balance over this control volume. Next, the pde is integrated over each control volume. The terms in the differential equations which are differentials over the space co-ordinate, i.e. the conduction terms, are evaluated using piece-wise continuous profiles. These profiles express the variation of the dependent variable between the grid points. Because of simplicity a linear piece-wise continuous profile is used.

Using the control volume method, the pde model equation 2.10 can be transformed for (N-2) grid points (excluding the boundaries), into (N-2) ode’s in the (N-2) temperatures at those grid points. In the derivations below, the nomenclature of figure 2.16 is used. Furthermore, the coordinate system is set up, such that, the centre of the pellet is at r=0 and the pellet surface at r=r_{pellet} and the subscript 'pellet' for pellet properties is omitted for clarity reasons.

\[
\rho c_p r^2 \frac{\partial T}{\partial t} = \lambda \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) \tag{2.20}
\]

\[
\rho c_p \int_0^r r^2 \frac{\partial T}{\partial t} \, dr = \int_0^1 \lambda \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) \, dr \tag{2.21}
\]

\[
\int_0^{r_p^3} \left[ \frac{1}{3} \rho c_p \frac{dT_p}{dt} \right] \, dr = r_p^2 \lambda \int_0^1 \left( T_E - T_P \right) \, dr - r_w^2 \lambda \int_0^1 \left( T_P - T_W \right) \, dr \tag{2.22}
\]

\[
\frac{dT_p}{dt} = \frac{1}{r_p^3} \rho c_p \left\{ \frac{r_p^2 \lambda (T_E - T_P)}{\partial r} - \frac{r_w^2 \lambda (T_P - T_W)}{\partial r} \right\} \tag{2.23}
\]
2.4.2 Boundary conditions

The above discretisation is valid for all the control volumes, except those at the boundaries. In the description of the boundary control volumes, the boundary conditions have to be used. This is done by defining those volumes as having half the size of a normal control volume (figure 2.16). A thermal energy balance is set up over the control volumes, in which the flows at one side of the volume follow directly from the boundary conditions.

2.4.2.1 Boundary condition at the centre of the pellet

The first boundary condition for equation 2.10 is that there is no heat flow going through the centre. The thermal balance over the control volume at the centre leads to the discretisation below.

\[
\frac{\rho \, c_p \, r^2 \, dT}{dt} = \frac{1}{dr} \left( \frac{d}{dr} \left[ 0 - r^2 (\lambda) \frac{\partial T}{\partial r} \right] \right)
\]

\[
\frac{1}{3} \frac{r^4 \, dT_C}{dt} = -r^2 (\lambda) \frac{\partial T}{\partial r}
\]

\[
\frac{1}{3} \frac{r^4 \, dT_C}{dt} = -r^2 \lambda (T_i - T_C)
\]

\[
\frac{dT_C}{dt} = \frac{1}{3} \frac{r^4 \, dT_C}{dt} \left( \frac{r^2 \lambda (T_i - T_C)}{\partial r} \right)
\]

2.4.2.2 Boundary condition at the outer surface of the pellet

For the pellet outer surface a similar analysis can be done. As was explained above, different models have been used to describe the flux at the pellet boundary. The simplest boundary condition is a constant surface temperature which results in a straightforward discretisation.

\[
\frac{dT_B}{dt} = 0
\]

The other two boundary conditions considered are radiation from the oven wall and a combination of radiation and conduction from the oven wall. For both boundary conditions it is first of all necessary to set up a thermal balance over the control volume at the pellet outer surface.

\[
\frac{\rho \, c_p \, r^2 \, dT}{dt} = \frac{1}{dr} \left( r^2 (\lambda) \frac{\partial T}{\partial r} \right) - \frac{\phi_q}{4\pi}
\]

\[
\frac{1}{3} \frac{r^3 \, dT_B}{dt} = r^2 (\lambda) \frac{\partial T}{\partial r}
\]

\[
\frac{1}{3} \frac{r^3 \, dT_B}{dt} = r^2 \lambda (T_i - T_B)
\]

\[
\frac{dT_B}{dt} = \frac{1}{3} \frac{r^3 \, dT_B}{dt} \left( \frac{r^2 \lambda (T_i - T_B)}{\partial r} - \frac{\phi_q}{4\pi} \right)
\]

By appointing to the flow at the boundary the appropriate equation, the discretisation for the outer boundary can be obtained. For a radiation boundary condition, equation 2.18 is used which leads to:
Numerical implementation

\[ \phi_q \Big|_B = \phi_{q,\text{radiation}} = \frac{4 \pi r_B^2 \sigma}{1 + \left(1 - \varepsilon + \frac{r_B^2}{r_{\text{oven}}^2} (1 - \varepsilon_{\text{oven}}) \right)} \left( T_{\text{oven}}^4 - T_B^4 \right) \]  
(2.33)

\[ \frac{dT_B}{dt} = \frac{1}{3 (r_B^2 - r_{\text{oven}}^2) \rho c_p} \left\{ \frac{r_B^2 \lambda_i (T_i - T_B)}{1 + \left(1 - \varepsilon + \frac{r_B^2}{r_{\text{oven}}^2} (1 - \varepsilon_{\text{oven}}) \right)} - \frac{r_B^2 \sigma}{1 + \left(1 - \varepsilon + \frac{r_B^2}{r_{\text{oven}}^2} (1 - \varepsilon_{\text{oven}}) \right)} \left( T_{\text{oven}}^4 - T_B^4 \right) \right\} \]  
(2.34)

For a combination of radiation and conduction at the boundary, the discretisation is obtained using equation 2.18 and 2.19:

\[ \phi_q \Big|_B = \phi_{q,\text{radiation}} + \phi_{q,\text{conduction}} \]  
(2.35)

\[ \phi_q \Big|_B = 4 \pi r_B^2 \frac{\sigma}{1 + \left(1 - \varepsilon + \frac{r_B^2}{r_{\text{oven}}^2} (1 - \varepsilon_{\text{oven}}) \right)} \left( T_{\text{oven}}^4 - T_B^4 \right) + \frac{\lambda_{\text{air}} r_{\text{oven}}}{(r_{\text{oven}} - r_B) r_B} \left( T_{\text{oven}} - T_B \right) \]  
(2.36)

\[ \frac{dT_B}{dt} = \frac{1}{3 (r_B^2 - r_{\text{oven}}^2) \rho c_p} \left\{ \frac{r_B^2 \lambda_i (T_i - T_B)}{1 + \left(1 - \varepsilon + \frac{r_B^2}{r_{\text{oven}}^2} (1 - \varepsilon_{\text{oven}}) \right)} - \frac{r_B^2 \sigma}{1 + \left(1 - \varepsilon + \frac{r_B^2}{r_{\text{oven}}^2} (1 - \varepsilon_{\text{oven}}) \right)} \left( T_{\text{oven}}^4 - T_B^4 \right) \right\} + \frac{r_B^2 \lambda_{\text{air}} r_{\text{oven}}}{(r_{\text{oven}} - r_B) r_B} \left( T_{\text{oven}} - T_B \right) \]  
(2.37)

2.4.3 Integration schemes

Through discretisation, the pde and the boundary conditions have been transformed into N ode's. Together with the initial conditions, these ode's have to be integrated over the required process time to give the numerical estimation of the solution of the pde. For the integration, a number of different schemes are available.

Accumulation and conduction term can be treated differently, but for both assumptions are needed as to how the temperature at the grid point varies with time. They can be dependent either on the grid point temperature at the beginning of the integration step (fully explicit), on the grid point temperature at the end of the integration step (fully implicit), a weighted average of both, or a weighted average including temperatures at previous integration steps.

Integration schemes based on the latter assumption are the Runge-Kutta and Gear schemes. Runge-Kutta, fourth-order with adaptive step-size control, is an accurate and reasonably stable scheme. Gear is less accurate but more apt to so called 'stiff differential equations'. The choice of integration scheme is dependent on accuracy, stability and calculation time. For the model with a constant surface temperature boundary condition the Runge-Kutta scheme is used. For the other two models, the Runge-Kutta does not perform well and a Gear scheme is used.

For more detailed information on integration schemes is referred to literature.

2.4.4 Fit procedure

The conductivity is the only undetermined parameter in each of the models describing the experiments. A minimisation method based on \( \chi^2 \) is used to determine the most likely value of the conductivity in each of the experiments. In \( \chi^2 \) the difference between the temperatures
measured in the experiment and the temperatures predicted by the model is evaluated. The $\chi^2$ function is defined by 28,

$$\chi^2 = \sum_{n=1}^{N} \left( \frac{T^{\text{exp}}(t_n) - T^{\text{mod}}(t_n)}{\sigma^{\text{exp}}(t_n)} \right)^2$$

(2.38)

wherein $N$ is the total number of times at which the pellet temperature was measured, $T^{\text{exp}}(t_n)$ the measured pellet temperature at the time of the $n^{th}$ measurement, $T^{\text{mod}}(t_n)$ the model prediction of the pellet temperature at the time of the $n^{th}$ measurement and $\sigma^{\text{exp}}(t_n)$ the standard error in the $n^{th}$ measurement of the pellet temperature.

The experimental standard error in each of the measured points can be estimated by two different methods. The error can be estimated from scatter in the measurements. This is done by fitting a small number of measurements around the measurement to a linear trend. The estimate error in the measurement is derived from the standard deviation from this trend. An other method is based on the published error of measurements with the thermocouple used as given in table 2.3.

The method used to find the value of the conductivity minimising $\chi^2$ for given experiment, is based on finding the minimum value within an given initial interval of the effective conductivity. This interval is chosen as $0<\lambda<2$ W/m-K; slightly larger than the interval, in which the conductivity is likely to be found according to figure 2.1 and 2.2. Using golden sections for fast convergence and parabolic interpolation for higher accuracy the interval in which the minimum is to be found is subsequently narrowed until a convergence criterion has been reached $24,26,27$. The convergence criterion has been set at 0.005 W/m-K.

When the value of $\chi^2$ is calculated for an experiment, the validity of the combination of model and value of the conductivity can be assessed 25. More precisely, the probability can be calculated, with which the experimentally found response will be measured when the process would be governed by the model proposed. This probability ($Q$) can be calculated using the relation,

$$Q = 1 - \frac{1}{\Gamma \left( \frac{N-M}{2} \right)} \int_0^{\chi^2 \frac{N-M}{2}} t^{(N-M)-1} e^{-t} dt$$

(2.39)

wherein $\Gamma$ is the gamma function, $N$ denotes the number of measurements in the experiment and $M$ the number of parameters determined (M). In the experiments only the conductivity is determined so $M$ is equal to one.

In general, for values of $Q$ smaller than 0.001, the model should be rejected. A value for $Q$ of 1 indicates that the model is correct within the error of the experiment. As can easily be seen, $Q$ is very sensitive to the error in the measurements.
2.5 Results and discussion

2.5.1 General remarks on the experimental results
In total 18 experiments have been conducted in the temperature range of 530 to 1350K. The intended temperature step of 50 K was in practice difficult to attain, so some scatter can be observed in the magnitude of the step-change (ΔT = 47±11K). The response curves are smooth and show very little scattering (figure 2.17). A clear trend of increasingly fast response with increasing temperature of the experiment is observed (figure 2.18).

2.5.2 Results with the constant surface temperature model
At first, the experimental response curves are analysed using the exact solution of the constant surface temperature model. Although this method might not be very accurate, depending on the conductivity, it provides quick and simple estimates for the conductivity in the different experiments.

For this method, the temperature response curves are transformed to a dimensionless temperature (θ) according to equation 2.9, in which the final, assumed equilibrium temperature of the response curve, is used as estimate of the surface temperature. As can be seen in figure 2.19, the experimental curves indeed show an approximate exponential behaviour, after a short time, as predicted by equation 2.11. The slope however, is only approximately a constant.

---

Figure 2.18, example of response curve

Figure 2.19, transformed experimental response curves
From the slope of a linear fit on the transformed response curve, the effective conductivity of the pellet is determined for every temperature interval using equation 2.11 and 2.1 (see figure 2.20 and appendix E).

An important factor influencing these results is the estimation of the surface temperature. The slope of the transformed response curve, and thus the conductivity estimate, is sensitive to small errors in the estimation of the surface temperature. The analysis is further improved by using the expected linearity of the transformed response curve to get a better estimate of the surface temperature. A surface temperature which is chosen too low or too high will cause the transformed response curve to significantly deviate from linearity, especially at the end. The surface temperature can be determined more accurately by requiring a linear trend even at the end of the response. Most response curves, after correction of the surface temperature, still show some deviation from a straight line. This could indicate that the assumption of a constant surface temperature is not valid. It could also be caused by the temperature dependence of the specific heat and the conductivity which are neglected.

From both the literature sources shown in figure 2.1-2.2 and the theoretical analysis, it is expected to find a conductivity between 0.4 and 1.2 W/m-K. The conductivities found in the present analysis are up to a factor 10 lower, especially at low temperatures. This difference together with the deviation from linearity mentioned above indicate that the assumption of constant surface temperature is not valid. This conclusion is supported by the fact that in the range of conductivities found, the resistance analysis shows that external resistance can not be neglected at higher temperatures and is dominant at the lower temperature (see figure 2.12). Omitting the external resistance in this situation will lead to significantly lower estimates of the conductivity as the response time caused by external resistance will be compensated for by increasing the response time of the internal conduction. To obtain more realistic values of the conductivity the external transport process should be incorporated in the model.

It should finally be noted, that according to the constant surface temperature theory, the slope of the curve is independent of the nature of the temperature measured. That is to say, the slope of the curve is independent of the position at which the temperature is measured and whether or not this temperature is an average temperature. The difference between the aforementioned curves is the time needed for the constant slope to begin. As can be concluded from figure 2.21, the measured temperature is not the temperature at the centre of the pellet, but a temperature somewhere in between the centre and the boundary. It is presumably an average temperature over a finite volume within the pellet.
Results and discussion

2.5.3 Results with a radiation boundary condition

From the results of the constant surface temperature analysis it is found necessary to include the external heat transport process. From the resistance analysis (figure 2.11) it is concluded that radiation is the dominant external transport process and will be used to model the external transport. Using such a model means using a numerical integration scheme according to equations 2.18-2.21 and 2.32, in combination with a minimisation scheme according to equation 2.36. These schemes are programmed in a Matlab environment. To verify the accuracy of the numerical scheme, the program is first run with a boundary condition according to equation 2.26. The results are in agreement with the exact solution to this problem (figure 2.22).

The physical influence of the boundary condition can be illustrated by the difference in internal temperature profile resulting from the difference between a constant surface temperature and a radiation boundary condition with $\lambda^{\text{eff}} = 0.2$ W/mK (figure 2.23).
The issue of which temperature is measured by the thermocouple is already mentioned in the constant surface temperature model analysis. In the present analysis, unlike in that model, this has a significant effect on the resulting estimate for the conductivity. From the constant surface analysis it is concluded that neither the average, nor the pellet centre temperature is measured. It is found in literature that a thermocouple measurement is especially sensitive to temperature gradients around its tip. The measured temperature is therefore most likely a volume average temperature over a finite part of the pellet around the tip. In the analysis it is assumed that this measured part can be described by a sphere concentric to the pellet but smaller. The influence of the size of the sphere lies in the beginning of the experiment; the smaller the sphere, the longer the initial time lag of the response (figure 2.24).
The size of the sphere has been determined by a best fit of the model to the apparent experimental time-lag. The resulting size of the sphere is 4/5 of the pellet radius from the centre (51% of its volume). This seems very large and will be further commented on in the section on the error in the thermocouple measurement.

The results of this model on the conductivity found are shown in figure 2.25 and further given in appendix E. The results of the constant temperature model are also included for comparison.

As expected, the conductivity found at high temperatures were similar, whereas at lower temperatures a much higher conductivity is found than with the constant surface temperature model. Apparently, the conclusion drawn from the constant surface temperature approach that the external transport was influencing the value of the conductivity is valid.

However, at lower temperatures another problem is encountered in the radiation model. The estimate of the conductivity rises sharply at the lower temperatures. This could be a physical reality were it not that for the experiments at the lowest temperatures no value for the conductivity can be found that would lead to a sufficiently fast temperature response. In other words, the fastest response, which is that of uniform heating, is slower than the experimentally found curve. This leads to the conclusion that the heat flow to the pellet is underestimated by the radiation model and additional external transport processes have to be included in the analysis.
2.5.4 Results with a combined radiation and conduction boundary condition

The resistance analysis of figure 2.11 show that the second most important external transport mechanism is conduction from the oven wall to the pellet surface. Therefore, to improve the modelling of the external transport, the external transport by conduction is included in the model. To incorporate this effect, the boundary condition in the numerical scheme of the radiation model, has been replaced by equation 2.35. The analysis with this model has lead to the following estimates of the conductivity (figure 2.19).

![Graph showing conductivity vs. temperature](image)

Figure 2.26, results of the analysis with both radiation and conduction

As expected the influence of the additional flow to the boundary is largest at the lowest temperatures. At higher temperatures the effect is a minor decrease in the estimate of the conductivity. The decrease is caused by the fact that the measured response time is partly explained by the conduction. The absence of this effect in the radiation model is compensated for by a higher conductivity estimate. Comparing both experimental conductivity curves this effect is not always observed. This is caused by the scattering influence of the minimisation scheme.

The effect of rapidly increasing conductivity at the lower temperatures is still visible. It is thought that this is still caused by the model of the external transport being insufficiently accurate. As at lower temperatures the external processes become more dominant, an increasingly detailed model is necessary for the external processes. An important aspect of the external transport process which has not been included in the model thus far is the temperature profile along the length of the oven. This means that the pellet is radiated on by a temperature which is significantly higher at one side than the other. In the model the pellet is assumed to be in an uniform temperature field whose magnitude is given by the experimentally measured equilibrium temperature. It could well be that the initial temperature response is strongly influenced by radiation from the hot part of the oven, resulting in a higher flow of heat than the one based on the equilibrium temperature. This in turn could explain the large estimates of the conductivity at lower temperatures.

The overall estimates of the conductivity found in the experiment and subsequent model analysis are still lower than those found in literature as can be seen in figure 2.27 and appendix E. This could be influenced by the accuracy of the procedure.
2.5.5 Accuracy of the estimation

2.5.5.1 Random errors

In the experiments, there are only a few sources available of random error. First of all, there will be a random error in the temperature measurement by the thermocouple. According to literature on thermocouple measurement, this is estimated on ±0.4%. At the measured temperature range this would lead to an error of 2-4°C. The manual of the imp states an error <0.1°C. The latter estimate is considered a better estimate as a similar error is observed in the scatter on the temperature response curves (0.01 < σ <0.1 see e.g. figure 2.18).

The position of the thermocouple within the pellet induces another random error. It is attempted to position the thermocouple in the centre of the pellet but there will be scattering. It is estimated that the error in the position is <1mm. The main influence of this error would be scattering in time lag between the step-change in temperature of the surroundings and the first response of the pellet. The response of the centre of the pellet will show a large time lag which is shortened when the position is more towards the outer surface. Another random error influencing the measured time lag in the experiment, is the finite time it takes to induce a step-change in surroundings temperature. This takes about 1-2 seconds and will also shorten the time-lag. It is very difficult to estimate what influence of this error has on the effective conductivity. However, as it was concluded that the thermocouple measures the average temperature over a large part of the pellet, it is not expected to have a large influence.

A fourth source of a random error is the shape of the pellet and its deviation from a spherical shape. As given in table 2.2, this amounts to only 1% of the diameter. When the pellet is rotated slightly with respect this would give an error but it can presumably be neglected. Finally slight changes in position or fluctuation in the oven heating during the experiment will result in random errors. These random errors are not likely to occur regularly but if they do occur they will very much influence the estimate of the conductivity.

Concluding it can be stated that the random errors are not very important in the experiments and are probably limited to the small error in the thermocouple measurement and errors in the time lag. The errors in the time lag can also be represented as errors in the thermocouple measurement, but than in the time co-ordinate instead of the temperature co-ordinate.

2.5.5.2 Validity of the model

When the random errors in the measured data are known, it can be analysed using equation 2.37, whether or not the combination of model and value of the conductivity is valid. The result of this validity check is the probability, that the measured response could have come
from a system with the characteristics of the model. For every measurement this probability is calculated using the model which includes both radiation and conduction at three different estimates of the random error. First the error is used which is calculated from the scattering in the response curve (0.1 K). Secondly two larger errors are used which could incorporate the time lag. The temperature in the response initially rises as fast as 0.5 K/s so the most likely estimate of the total random error would be 0.3 K (0.2 K above the 0.1 K scatter in the temperature). A more pessimistic estimate is 0.5 K. The results of the analyses are given in table 2.7.

<table>
<thead>
<tr>
<th>Table 2.7, Statistical probability of the modelled curve for different values of the error</th>
<th>temperature of the experiment (K)</th>
<th>error (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>554</td>
<td>605</td>
</tr>
<tr>
<td>&lt;0.1</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>0.3</td>
<td>0.71</td>
<td>0</td>
</tr>
<tr>
<td>0.5</td>
<td>1</td>
<td>1</td>
</tr>
</tbody>
</table>

It is immediately clear from the results that the scatter in the temperature measure alone is not enough to ensure the validity of the model used as they all prove invalid with this error (Q=0). Furthermore, an error of 0.5 K will result in the model being valid for all the experiments. At the best estimate of the error, 0.3 K, gives a scattered picture. This could either mean that the estimation of the random error is too small, or it could indicate that the model is not valid. The use of an invalid model is a form of systematic error. In the next section it will be analysed what the influence of statistical errors is on the conductivity determined.

2.5.5.3 Sensitivity analysis
To estimate the influence of systematic errors on the accuracy of conductivity results, the sensitivity of the model result has been determined for all parameters in the model. The sensitivity analysis has been performed by determining the change in the estimate of the conductivity when a perturbation of +5% was introduced on a model parameter. A 5% deviation in the value of the parameters is reasonable although in most cases pessimistic. This has been done for experiments at two different temperatures an the results are given in table 2.5-2.6.

<table>
<thead>
<tr>
<th>Table 2.5, Result of sensitivity analysis on experiment at low temperature</th>
</tr>
</thead>
<tbody>
<tr>
<td>process conditions of experiment: $T_{p}= 742 K$, $T_{oven}= 794 K$, $\lambda_{eff}=0.31$, W/m·K</td>
</tr>
<tr>
<td>variable which is changed</td>
</tr>
<tr>
<td>---------------------------</td>
</tr>
<tr>
<td>pellet diameter</td>
</tr>
<tr>
<td>pellet emissivity</td>
</tr>
<tr>
<td>pellet density</td>
</tr>
<tr>
<td>pellet specific heat</td>
</tr>
<tr>
<td>oven reflectivity</td>
</tr>
<tr>
<td>measured temperature volume ($0.7*r_{pellet}$)</td>
</tr>
<tr>
<td>measured temperature volume ($0.9*r_{pellet}$)</td>
</tr>
</tbody>
</table>

From the sensitivity analysis it can be conclude that the error in the conductivity lies in the range of 20%. This magnitude of error is supported by the apparent scatter in the curve of conductivity temperature dependency between temperatures of 700 to 1300 K as shown in figure 2.27. Both at higher and lower temperatures this error is not sufficient to explain the scatter.
Table 2.6, Result of sensitivity analysis on experiment at high temperature process conditions of experiment: \( T_0 = 1214 \text{K}, T_{\text{oven}} = 1247 \text{K}, \lambda_{\text{eff}} = 0.43 \text{ W/m-K} \)

<table>
<thead>
<tr>
<th>variable which is changed</th>
<th>change in conductivity estimate</th>
</tr>
</thead>
<tbody>
<tr>
<td>+5%</td>
<td></td>
</tr>
<tr>
<td>-2%</td>
<td></td>
</tr>
<tr>
<td>-2%</td>
<td></td>
</tr>
<tr>
<td>+9%</td>
<td></td>
</tr>
<tr>
<td>+9%</td>
<td></td>
</tr>
<tr>
<td>-2%</td>
<td></td>
</tr>
<tr>
<td>+19%</td>
<td></td>
</tr>
<tr>
<td>-19%</td>
<td></td>
</tr>
</tbody>
</table>

Regarding the error in the conductivity at the highest temperature measured (1341 K), it is expected to be larger as the step change in that experiment was significantly lower than in the other experiments (viz. 16 K). This could explain the significantly higher value obtained in comparison with temperatures slightly lower. A small temperature step has a negative effect on the relative accuracy of the temperature changes measured. The fast response time at higher temperatures poses an additional negative influence as the effect of a non-ideal step change becomes relatively more important. At lower temperatures (<700K) the simple model of the external heat transfer is likely to influence the estimate of the conductivity as was explained previously.

All in all, it is evident that the apparent error in the estimation of the conductivity due to the for mentioned systematic errors in the model (±20%) cannot account for the large difference between the measurements and literature (>100%) given in figure 2.19. Before it can be concluded that this difference found is a physical reality, the systematic errors in the temperature measurement has to be looked upon.

2.5.5.4 Errors in the temperature measurement method

The temperature error mentioned in the analysis on the validity of the model, refers to the difference in read out of the temperature and the actual temperature at the thermocouple junction. In this section it is analysed whether the temperature of the thermocouple junction is actually the same as the temperature of the pellet. These can differ significantly due to heat losses/gains through the thermocouple wire.

Most likely heat losses are due to the temperature difference between the hot and cold thermocouple junction which are at the temperature of the pellet and the room temperature respectively. This effect is estimated first by a theoretical approach, calculating the resistance of the entire length of the thermocouple combined with the difference between the pellet temperature and room temperature. This flow is also estimated using the temperatures measured in the insulation by an additional thermocouple (included in figure 2.5). In this estimation the resistance of the thermocouple wire between the insulation and the tip is calculated and combined with the measured temperature difference. The thermocouple can gain heat through direct radiation on the small part of the thermocouple between the insulation and the pellet surface which is unprotected. This heat input is calculated using equation 2.3 in which the pellet radius is changed to the thermocouple radius.

These heat effects of radiation and conduction are added and compared to the input of heat at the pellet surface the analysis of which is given in appendix D. The resultant net heat loss percentage is dependent on the temperature at which the experiment is conducted and can be found in figure 2.28. In this figure the results of approach using the experimental temperature in the insulation is denoted ‘thermocouple’ and the approach using the temperature difference between pellet and room temperature is denoted 'theoretical'. It can be seen in figure 2.28 that in both analyses the net heat flow to the thermocouple through the wire is very limited compared to the heat flow reaching the pellet surface.
The second possible error in the temperature measurement is associated with the assumption of the temperature measured being an volume average over a part of the pellet volume. The fact that in order to predict the initial time-lag measured an average temperature over 50% of the pellet volume is needed seems large. This could indicate that the thermocouple wire is acting as a shortcut for heat transport within the pellet. Another possibility is the influence of the random errors of thermocouple location and non ideal step-change. Although these errors are considered random the effect will always be a shortening of the measured time lag in the response. As can be seen in figure 2.24 a slight decrease in time-lag will have a large impact on the size of the volume over which the average temperature is measured.
2.6 Conclusions and recommendations

From the experiments the effective conductivity of a single hematite pellet has been determined; 0.3 W/mK at 700K which rises with increasing temperature to 0.7 W/mK at 1400K with an accuracy of around 20%. Compared with the available literature on the conductivity this is relatively low which cannot be accounted for by the error. The reason for this might be the fact that the pellet was not adequately sintered. According to Biswas\textsuperscript{27}, the complete sintering of hematite needs temperatures as high as 1600K, while the pellet was sintered at approximately 1400K. When the pellet is not adequately sintered the interconnection of the ore particles in the pellet is less and thus the conductivity will be lower. In this case however, a stronger increase conductivity with rising temperatures would be expected although it is frequently very difficult to predict the exact behaviour of porous materials due to the combined and related effects of solid and fluid conductivity in very small pores\textsuperscript{18}.

At temperatures below 700K the conductivity seems to rise sharply but the accuracy of these conductivities is thought to be much lower, as the influence of external heat transport increases which is not modelled in enough detail. It is thought that the external heat transport is underestimated and thus a higher conductivity is found. In order to improve the accuracy of the analysis in general and especially at lower temperatures, the following recommendations are made. The modelling of the external process can be improved by incorporating a better estimation of the geometry of the experimental set-up. In the present analysis the geometry of a pellet in a tube is approximated by two concentric spheres; this should be replaced by the geometry of a sphere within a coaxial cylinder. In literature relations have been found to implement this for the model including radiation\textsuperscript{28,29}. In principle this should also be possible for the model including both conduction and radiation. Furthermore, the temperature gradient along the length of the oven should be modelled as well. This can be approximated for the radiation by dividing the tube into small rings of uniform temperature from which the radiation to the pellet surface can be calculated\textsuperscript{28}. In the model referred, pellet surface has to be considered uniform in temperature. Introducing these adjustments in the present model is relatively simple as the model of the pellet remains one dimensional. With the introduction of even more detail the model is likely to become two or three dimensional. Then the use of a CFD model could be considered.

An other major improvement can be made in the knowledge on the error of the temperature measured. It has been necessary to assume that the temperature measured is an volume average temperature over the innermost 50% of the pellet which could be caused by conductive effects of the thermocouple. In order to verify this, an extra thermocouple could be inserted off centre. With a second measurement it will be possible to use the differential of the temperature as variable. Finally, it should be attempted to insulate the part of the thermocouple wire just of the tip which is also in contact with the pellet.

Although these recommended improvements would likely increase the accuracy of the estimated effective conductivity, the method in general is not ideally suited for the determination of the conductivity as a step-change on the pellet surface temperature is required. With the tube oven as available means, only the environment of the pellet surface can be changed. In that case it is necessary for the external heat transport resistance to be negligible compared to the internal heat transport resistance. This is not valid for the tube oven which results in the necessity of complicated models which do not improve the accuracy of the experiment.

It is recommended to use an experimental set-up in which the heat is transported to the pellet by a more conductive medium than air for example oil. The step change can for example be induced by submerging the pellet in an isothermal, well stirred vessel filled with oil. This would, however, not be very straightforward, as a volume of oil at the temperatures necessary (up to 1300 K) would be a serious safety hazard.
Although the experiment has not been ideal, it has given a large amount of insight into the mechanisms of heat transport. As the tube oven is also used to conduct other experiments at Hoogovens this insight could contribute in the analysis of those experiments. Furthermore, the experience gained in the method of analysis has proven to be of worth in the analysis of the experiments on the packed bed, as similar analyses were necessary.
2.7 List of symbols

- **A** surface area \([m^2]\)
- **c_p** specific heat \([J/kg-K]\)
- **d** diameter \([m]\)
- **g** gravitational acceleration \((9.81)\) \([m/s^2]\)
- **Gr** Graetz number \((x^2g\Delta\rho\eta^2)\) \([-]\)
- **h** heat transfer coefficient \([W/m^2-K]\)
- **M** number of parameters varied \([-]\)
- **N** number of measured points in experiment \([-]\)
- **Nu** Nusselt number \((h\cdot d/\lambda)\) \([-]\)
- **Pr** Prandt number \((c_p\cdot\rho\cdot\eta)\) \([-]\)
- **Q** Probability \([-]\)
- **R** resistance to heat flow \([m^2\cdot K/W]\)
- **T** temperature \([K]\)
- **\langle T \rangle** volume average temperature \([K]\)
- **t** time \([s]\)
- **r** radius/radial position \([m]\)
- **V** volume \([m^3]\)
- **\chi^2** chi squared \([-]\)
- **\delta r** radial difference \([m]\)
- **\varepsilon** emissivity \([-]\)
- **\phi_q** heat flow \([J/s]\)
- **\eta** dynamic viscosity \([Pa\cdot s]\)
- **\lambda** conductivity \([W/m-K]\)
- **\rho** density \([kg/m^3]\)
- **\sigma** Stefan-Boltzman constant \((5.67\cdot 10^{-8})\) \([W/m^2-K^4]\)
- **\sigma** standard error \([-]\)
- **\tau** characteristic time \([s]\)
- **\Gamma** gamma function \([-]\)
- **\Theta** dimensionless temperature \([-]\)

**sub and superscripts**

- **air** of the air
- **avg** average
- **conduction** through the mechanism of conduction
- **convection** through the mechanism of convection
- **eff** effective
- **exp** experimentally measured
- **external** externally of the pellet
- **free conv** through the mechanism of free convection
- **internal** internally of the pellet
- **mod** calculated by model
- **n** number of measured point
- **oven** at the oven wall
- **pellet** of the pellet
- **plate** of a flat plate
- **pore** of the pores
- **radiation** through the mechanism of radiation

- **B** at grid point on the outer boundary
- **C** at grid point on the centre boundary
- **E** at grid point east of grid point P
- **e** at the interface between the control volumes of grid points P and E
- **I** at the last grid point before the boundary
- **i** at the interface between the control volumes of grid points I and B
List of symbols

P .................. at the grid point under consideration
W .................. at the grid point west of grid point P
w .................. at the interface between the control volumes of grid points P and W
0 .................. at the beginning of the experiment (t=0)
2.8 References

3. Heat transport in the packed bed

3.1 Introduction and objective

This chapter consists of an investigation into the heat transport within a packed bed of pellets heated by a flow of hot gas at the conditions found in the firing section of the pellet production process. The direct objective of the investigations is to quantify the heat transfer from gas to the pellets and its dependency on three important process variables viz., temperature, flow rate and bed voidage. For more detailed information on the choice of these variables is referred to chapter one.

The process of heat transport in packed bed under the conditions mentioned, is highly complex and consists of both transport to the surface of the pellets and transport within the pellets. These two heat transport processes will subsequently be referred to as, external and internal respectively. As the internal heat transport has been investigated in the previous chapter, the emphasis will be on the external heat transport. In experiments only the combination of both transport processes can be measured. In order to find the effect of the external heat transport, the internal transport has to be corrected for, using the results from the investigation of the previous chapter.

External heat transport can be accomplished either directly from the fluid phase through conduction or convection, or indirectly from the surface of neighbouring pellets by conduction or radiation. Adding to the complexity of the problem is the disordered structure of the bed. In order to overcome this complexity in describing the system, the different mechanisms of heat transport from the fluid to the pellet can be lumped into one (effective) interfacial transport term. This term has the form of Newton's law of cooling, as the interfacial heat flow \( \Phi_{\text{eff}} \) is related to the driving force given by the difference in the bulk fluid temperature \( \langle T \rangle^f \) and the temperature of the solid \( \langle T \rangle^s \), by the interfacial heat transfer coefficient \( h_{\text{eff}} \) per unit of interfacial area \( A_{\text{eff}} \). The effects of the different mechanism are thus lumped into the heat transfer coefficient. As a result of this lumping, the values and trend of this coefficient will be strongly influenced by the set-up of the experiments, e.g. combination of solid and fluid and shape of the solid phase. This is reflected in the results of former investigations on the heating of packed bed by a hot fluid as can be seen in figure 3.1.

In general, very little attention has been given in literature to the description of the heat transfer coefficient in the pellet firing process. Most attention has been given to heat transfer in packed beds as used in reactors, which are either heated or cooled by the wall. In this situation, the emphasis is on the radial heat transfer, whereas in the pellet firing process the axial heat transport is most important. Furthermore, the description of the influence on the heat transfer of decreasing bed voidage through the presence of fines, has mostly been limited to the use of an average particle diameter. It is thought that the influence of fines on the flow and heat transfer is underestimated in this way, especially when there is a large difference in size between the particles and the fines.

The aforementioned lack of literature data has been the incentive of conducting experiments to determine the interfacial heat transfer coefficient for the pellet firing process and its dependency on temperature, gas flow rate and bed voidage. To this end, a pilot scale experimental set-up of the induration process was used, consisting of a cylindrical packed bed which could be heated by permeating flow of gas at elevated temperature, while measuring the temperature at different positions in the bed.

Although different types of experiments can be conducted to determine the heat transfer coefficient of a packed bed\(^2\text{-}\text{3,4}\), the available experimental set-up was most suited to

Heat transport in the pellet firing process
measuring the transient response of the bed, initially at uniform temperature, to a step change in gas inlet temperature. For these type of experiments relatively simple models exists by which the heat transfer coefficient can be determined from the response of the bed.

In these simple models a radial uniform temperature is assumed. Therefore, a major difficulty in the analysis of the experiments has been the presence of unexpected high heat loss to the wall containing the packed bed. Furthermore, a rising inlet gas temperature was encountered which is also not accounted for in the simple models. Although this has severely limited the analysis of the experiments, different models have been developed in order to try and overcome these difficulties, ultimately leading to a comprehensive numerical model including the transport within the wall.

![Graphs showing interfacial heat transfer coefficient dependency on temperature and flow in the pellet firing process according to different models: (A) Wakao and Kaguei: \( N_{up} = 2 + 1.1 \cdot \text{Re}_{p}^{0.6} \cdot \text{Pr}^{0.33} \), (B) Ranz and Marshall: \( N_{up} = 2 + 1.8 \cdot \text{Re}_{p}^{0.5} \cdot \text{Pr}^{0.33} \), (C) Kitaev: \( h_v = 114 \cdot w_{0.7} \cdot T^{3/2} \cdot \eta_{0.9} \), and \( f(\varepsilon) = 1.53 \cdot \varepsilon^{0.29} / \varepsilon^{0.36} \).]

Figure 3.1a-d, literature findings for the dependency of the interfacial heat transfer coefficient on temperature and flow in the pellet firing process according to:

(A) Wakao and Kaguei: \( N_{up} = 2 + 1.1 \cdot \text{Re}_{p}^{0.6} \cdot \text{Pr}^{0.33} \)

(B) Ranz and Marshall: \( N_{up} = 2 + 1.8 \cdot \text{Re}_{p}^{0.5} \cdot \text{Pr}^{0.33} \)

(C) Kitaev: \( h_v = 114 \cdot w_{0.7} \cdot T^{3/2} \cdot \eta_{0.9} \cdot f(\varepsilon) \) and \( f(\varepsilon) = 1.53 \cdot \varepsilon^{0.29} / \varepsilon^{0.36} \)
3.2 Experimental

3.2.1 Experimental set-up

The experiments on the packed bed were conducted using the 'Pellet Proef Installation' (PPI) at Hoogovens. The PPI is a set-up for pilot scale experiments on the pellet induration process. The PPI consists of a cylindrical shaped pot that contains the packed bed, a gas burner producing a flow of hot gas which is led through the bed via an exhaust to the stack. The set up is shown in figure 3.2 where the gas flow is from top to bottom.

![Figure 3.2, experimental pilot scale set up (PPI)](image)

3.2.1.1 Burner section

In the burner section four different gas inputs are located: natural gas, primary combustion air, secondary combustion air and oxygen. The gas and primary combustion air are fed directly to the burner head in stoichiometric volume ratio (1:10), attained by a ratio control. The secondary combustion air is used to quench the primary combustion products to the desired temperature and flow. The oxygen input is not used in the experiments.

In the experiments a constant temperature and flow through the pellet bed is attained by flow control on both the natural gas flow and the secondary combustion air flow, as it is faster and more stable than direct control on temperature and total flow rate. It has to be noted that all flow rates mentioned here are normal volumetric flow rates, which is the volume flow rate evaluated at 273K. The temperature set points for each combination of total flow rate and temperature can be estimated by extrapolation from a first experiment at each value of the total flow rate. In this method, constant heat of combustion, density and specific heat of the gas are assumed. With this assumption, the resultant temperature \( T \) for different ratios of natural gas flow rate \( \Phi_v,\text{gas} \) and secondary combustion air flow rate \( \Phi_v,\text{secnd} \) can be estimated by

\[
T = \Lambda \cdot \frac{\Phi_v,\text{gas}}{\Phi_v,\text{total}}
\]  

(3.2)

\[
\Phi_v,\text{total} = \Phi_v,\text{gas} + \Phi_v,\text{prim} + \Phi_v,\text{secnd}
\]  

(3.3)

where \( \Phi_v,\text{total} \) denotes the total flow rate and \( \Phi_v,\text{prim} \) the primary combustion air flow rate. The constant \( \Lambda \) is determined for every value of the total flow rate (e.g. 3.8-10^4 K for 279 Nm^3/hr).
Using the flow control mentioned above, it takes approximately 30s to complete a step change in temperature, after which the total flow rate is stable within a deviation of ±5 Nm³/hr. The scattering in the gas inlet temperature thus induced, is dependent on the total flow rate and ranges from ±2 K to ±10 K. An important drawback of the flow control is the fact that the gas inlet temperature does not stay constant, but shows a continuous increase during the experiments of approximately 0.5 K/min as can be seen in figure 3.3, which is probably caused by the heating up of the burner section.

Figure 3.3, example of inlet and outlet gas temperatures during an experiment

3.2.1.2 Packed bed section
The pot containing the packed bed of pellets, is supported at the bottom by an iron grate consisting of grate bars of 2 cm in diameter which can be seen in figure 3.4. The inner surface of the pot is curved in a vertical wave pattern. This is to prevent extreme flows along the wall of the pot during an experiment induced by heat losses and an increased bed voidage at the wall. To minimise the heat losses, the wall of the pot is insulated using three layers of material of different density and conductivity which are given in table 3.1.

Figure 3.4, schematic picture of the pot

Heat transport in the packed bed
Even though the pot wall was insulated, it was expected that some heat losses would occur due to the heating up of the material of the wall. However, as it turned out, the heat losses are significant compared to the effect that is being measured in an experiment and can be as high as 70% over one experiment as will be shown in the results section. This has influenced the results significantly.

Table 3.1, properties of insulation material

<table>
<thead>
<tr>
<th>properties</th>
<th>Silimanit</th>
<th>IFF-1111</th>
<th>Superex 1900</th>
</tr>
</thead>
<tbody>
<tr>
<td>density (kg/m³)</td>
<td>2500</td>
<td>2100</td>
<td>2100</td>
</tr>
<tr>
<td>conductivity (W/m-K)</td>
<td>2.5</td>
<td>0.3</td>
<td>0.15</td>
</tr>
<tr>
<td>specific heat (J/kg-K)</td>
<td>1000</td>
<td>1000</td>
<td>1000</td>
</tr>
</tbody>
</table>

3.2.2 Experiments

In the experiments the response of the bed was observed to a step change of approximately 50K in the gas inlet temperature using the flow controls on the different gas input streams. This was done at different conditions of the initial gas temperature, the flow and the bed voidage as given in table 3.2. The flow rates given are the set points used in the control of the PPI; the actual measured flow rates were somewhat lower. It is assumed that over the temperature range of 50 K the heat transfer coefficient can be taken as a constant in order to determine a temperature dependency. The flows and temperatures are chosen to resemble the operating conditions of pellet production as given in chapter 1. The flow conditions used, which can be found in table 3.3, result in a Reynolds number based on the pellet diameter in the range of 450–2400. The range of process conditions is limited by the maximum temperature of the grate bars (900 °C) and the maximum pressure drop over the bed (870 mm H₂O). Due to the limited pressure drop, only the lower flow rates could be tested with experiments at lower voidage.

Table 3.2, experimental conditions

<table>
<thead>
<tr>
<th>variable</th>
<th>range</th>
</tr>
</thead>
<tbody>
<tr>
<td>temperature (°C)</td>
<td>350 – 900</td>
</tr>
<tr>
<td>bed voidage (-)</td>
<td>0.41</td>
</tr>
<tr>
<td>gas flow rate (Nm³/hr)†</td>
<td>150, 279, 425, 558, 700</td>
</tr>
</tbody>
</table>

Table 3.3, flow conditions in the experiments

<table>
<thead>
<tr>
<th>gas flow rate (Nm³/hr)</th>
<th>( V_{\text{superficial}} ) (Nm/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>150</td>
<td>0.5</td>
</tr>
<tr>
<td>279</td>
<td>0.9*</td>
</tr>
<tr>
<td>425</td>
<td>1.3</td>
</tr>
<tr>
<td>558</td>
<td>1.7</td>
</tr>
<tr>
<td>700</td>
<td>2.1</td>
</tr>
</tbody>
</table>

† average situation in the pellet production

In the standard use of the pot, it is filled with approximately 85 kg of pellets which results in a bed height of approximately 0.48 m. In the experiments, indurated pellets are used, which are sieved to obtain a size range 11-13 mm. The indurated pellets consist solely of hematite (Fe₂O₃), as not to influence the heat transport by the effects of evaporation, oxidation and sintering. The pellet properties are summarised in table 3.4. A packed bed of these pellets has an approximate voidage of 0.41. The bed voidage was changed using fines in the size range 1-3 mm which were obtained from grinding the pellet material (see table 3.5). This size range was chosen as it was small enough to significantly change the bed voidage without

† Nm³/hr is the volume flow rate at 273K

Heat transport in the packed bed

46
radically changing the total mass of the bed and large enough not be carried away with the gas flow.

Table 3.4, pellet properties

<table>
<thead>
<tr>
<th>material</th>
<th>Indurated pellets:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe₂O₃ (99.7wt%)</td>
<td></td>
</tr>
<tr>
<td>outer diameter (mm)</td>
<td>12±1</td>
</tr>
<tr>
<td>density (kg/m³)</td>
<td>3280 ±100</td>
</tr>
<tr>
<td>specific heat (J/kg-K)</td>
<td>800-1000</td>
</tr>
</tbody>
</table>

Table 3.4, bed conditions

<table>
<thead>
<tr>
<th>properties</th>
<th>voidage 0.41</th>
<th>voidage 0.39</th>
<th>voidage 0.37</th>
</tr>
</thead>
<tbody>
<tr>
<td>bed height (m)</td>
<td>0.48</td>
<td>0.49</td>
<td>0.49</td>
</tr>
<tr>
<td>mass pellets (kg)</td>
<td>85.0</td>
<td>84.3</td>
<td>85.0</td>
</tr>
<tr>
<td>mass fines (kg)</td>
<td>-</td>
<td>4.50</td>
<td>7.85</td>
</tr>
<tr>
<td>(d_{max}) (mm)</td>
<td>-</td>
<td>2.0</td>
<td>2.1</td>
</tr>
<tr>
<td>size distribution fines</td>
<td>-</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>&gt;3.15 mm (wt%)</td>
<td>-</td>
<td>54</td>
<td>57</td>
</tr>
<tr>
<td>&gt;2.0 mm (wt%)</td>
<td>-</td>
<td>43</td>
<td>40</td>
</tr>
<tr>
<td>&gt;1.0 mm (wt%)</td>
<td>-</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>&gt;0.5 mm (wt%)</td>
<td>-</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3.2.3 Measurement method

In the experiments, the response of the bed to a step change in gas inlet temperature was measured using thermocouples in the bed. As will be discussed in detail below, the temperature in, above and underneath the bed was measured using several different techniques including inserting thermocouples in pellets to overcome the difficulty of determining exactly which temperature is measured.

The standard means of temperature measurement in the PPI is by six thermocouples positioned at the centre of the pot at different heights as shown in figure 3.3 (right side of the pot). These thermocouples are R/S-type (Pt-Rh, d=1 mm) and protected by a ceramic tube (d=1 cm). The temperature of the thermocouples is recorded with a frequency of 0.1 Hz and a precision of 1 °C by the process control system of the PPI. The published error of this type of thermocouple is given in table 3.5.
Apart from the standard thermocouples, seven extra K-type (Ni-CrNi) thermocouples are used within the bed in the experiments, of which some are shown in figure 3.5 on left side of the pot. Four of these thermocouples with a diameter of 3 mm were inserted into the centre of a pellet. The remaining unprotected wire was insulated using ceramic tubes of the same size and material as the standard thermocouples. The space between the ceramic tube and the pellet surface was sealed using the same material as the pellet. These thermocouples, shown in figure 3.6, were positioned at different levels in the bed, at the same radial position and height as some standard thermocouples (viz. 11, 20, 40.5 and 50 cm from the top of the pot).

Two other thermocouples were positioned at 11 cm; one of them in the same way as the above mentioned four, the only difference being the size of the thermocouple (d=1 mm), the other thermocouple being completely unprotected. The different thermocouples at 11 cm in the bed are shown in figure 3.7. The last thermocouple was positioned in the top layer of the bed and was protected in the same way as the standard thermocouples.

The temperature of these seven thermocouples was recorded with a frequency of 0.1 Hz and a precision of 0.1°C using an isolated measuring pod (imp) connected to a personal computer. The imp measured a signal of ±12V and used a 12 bits AD converter. Its specified sensitivity is <0.1°C for the type of thermocouple used. The published error of a K-type thermocouple measurement is given in table 3.5.
Finally, both the gas temperature above the top and under the bottom of the bed are measured using a suction pyrometer which is positioned in the centre of the gas flow. In the suction pyrometer the temperature is measured using a R/S type thermocouple at a frequency of 0.1 Hz and with a precision of 1 °C by the process control system. The flow rate of the suction pyrometer was 13 Nm³/hr and the reported accuracy 2% on the temperature measured.

3.2.3.1 Thermocouple measurement
In the experimental set up, the above mentioned different thermocouples are used within the bed. The reason for this is that it is very difficult to accurately assess how the measured temperature relates to the temperatures of phases in the bed. A thermocouple interacts with its surroundings by heat transfer and the temperature measured is at best an expression of the equilibrium reached. Even the measurement of a suction pyrometer, which in general can be thought to measure the gas temperature can be influenced by radiation on the pyrometer tube. The thermocouples in the bed are even more sensitive to external influences.

For an unprotected thermocouple inserted in the bed, heat is transferred to the thermocouple through convection from the fluid and conduction from the fluid phase, and radiation and conduction from the solid phase. The exact result is impossible to predict apart from the fact that the measured temperature will be somewhere in between the solid phase surface temperature and the gas temperature. It is illustrative that it is sometimes stated that in this case, the measured temperature will be very close to the gas temperature, whereas in other research it is stated that the temperature measured will be the pellet surface temperature. The opinion in this project has been that the latter assumption will be more valid, however the situation might very well depend on whether or not, and how the thermocouple is protected. In a first approximation, the measurement of the standard thermocouple has been regarded the average temperature of the pellet as will be further discussed in the results.

In a new approach to overcome the aforementioned problem, the tip of the thermocouple has been inserted in a pellet. Effects by interaction of the thermocouple with gas phase are minimised by insulating the rest of the thermocouple with a ceramic tube and by sealing the area between the ceramic insulation and the pellet with the same material as the pellet. In this case, it can be assumed that the thermocouples measures the temperature of the pellet. As the thermocouple is relatively large compared to the diameter of the pellet, viz. 3 mm and 12 mm, it can be further assumed that the measured temperature is an average temperature of the pellet.

<table>
<thead>
<tr>
<th>Table 3.5, Thermocouple accuracy</th>
<th>R/S (Pt-Rh)</th>
<th>K (NiCr-Ni)</th>
</tr>
</thead>
<tbody>
<tr>
<td>limits of error (based on °C)</td>
<td>±0.1%</td>
<td>±0.4%</td>
</tr>
</tbody>
</table>
3.3 Theory

3.3.1 Interfacial heat transfer coefficient

In the most simple model of a packed bed, local thermal equilibrium between the phases is assumed. However, when the temperature at the bounding surfaces of the two phases changes significantly with respect to time, and when solid and fluid phases have significantly different heat capacities and thermal conductivities, the local rate of change of temperature for the two phases can not be assumed equal. This is the case in the firing experiments and a two phase model of the packed bed is likely to be necessary.

The heat transport dynamics of a packed bed, in a non equilibrium situation, can be described by a two phase model in which the local volume averaged properties of both solid (s) and fluid phase (f) are used to overcome the complexity of the bed structure. This leads to the coupling between the energy equation for each phase as formulated in compact form by Carbonell and Whitaker in which a total convective velocity vector is \( \mathbf{v} \), with components \( v^s, v^f, \), \( v^ss \) and \( v^sf \) is defined such that,

\[
\frac{\partial \langle T \rangle^f}{\partial t} + \mathbf{v}^f \cdot \nabla \langle T \rangle^f + \mathbf{v}^s \cdot \nabla \langle T \rangle^s = \nabla \cdot \mathbf{D}^f \cdot \nabla \langle T \rangle^f + \nabla \cdot \mathbf{D}^s \cdot \nabla \langle T \rangle^s + \frac{\mathbf{A}^s}{\rho c_p} h^s (\langle T \rangle^s - \langle T \rangle^f) \\
\frac{\partial \langle T \rangle^s}{\partial t} + \mathbf{v}^s \cdot \nabla \langle T \rangle^s + \mathbf{v}^sf \cdot \nabla \langle T \rangle^f = \nabla \cdot \mathbf{D}^f \cdot \nabla \langle T \rangle^s + \nabla \cdot \mathbf{D}^s \cdot \nabla \langle T \rangle^s + \frac{\mathbf{A}^f}{\rho c_p} h^f (\langle T \rangle^f - \langle T \rangle^s)
\]

(3.4), (3.5)

where \( t \) denotes time, \( V \) the volume, \( \rho \) the density, \( c_p \) specific heat and \( D \) the total thermal diffusivity tensors. The diffusivity tensors describe the influence, both within and between the phases, of conduction, hydrodynamic dispersion and radiation. The equations were in fact developed for the transient heat transfer in a packed bed with a steady flow.

As was shown in equation 3.1, the interfacial heat transfer coefficient is used as an overall convective heat transfer from the fluid to the solid phase, in which many characteristics of the system under consideration are lumped. When this heat transfer coefficient is determined experimentally, it is important to note whether or not, all the terms of equation 3.4 and 3.5 have been included. The use of (over)simplified versions results in the inclusion of the neglected terms into the heat transfer coefficient and therefore, in values for this coefficient that are valid only for those particular experiments.

It should be further noted, that in general, as the thermal conductivity of the solids is not large enough to lead to an isothermal solid temperature, the conductivity of the solid also influences the temperature field around it. Therefore, the convective heat transfer coefficient obtained from a given fluid-solid combination is not expected to be valid for some other combinations.

Although using equation 3.4 and 3.5 to analyse experiments has the advantage of finding a heat transfer coefficient that is independent of the experimental set-up, it is not practically feasible due to the difficult modelling and the large number of variables that need to be measured or approximated. Therefore, more simple models are used in which terms are omitted and thus influences lumped in the heat transfer coefficient. For example, in the experiments conducted in this project only the central temperature of the bed was measured thus no information exists on the radial temperature distribution making it not desirable to include radial transport within the model. The models which can be used to predict the experiments will therefore have only an axial dependency and will thus be one dimensional. This is no principle problem, as long as the heat transfer coefficient obtained from the pilot scale experiments is also valid for the pellet production process.
3.3.2 Quasi single-phase model

A very simple model of a packed bed is the quasi single-phase model. It is a quick and simple method to get estimates of the heat transfer coefficient. The quasi single-phase model is derived from looking at the top layer of the bed as can be seen in figure 3.8 in which the effect of heat losses can effectively be discarded. It is quasi single-phase, as it is assumed that the particles in the first layer of the bed have a negligible effect on the fluid temperature. It is further assumed that all the particles in the layer have the same, uniform temperature, are spherical of shape and have thermophysical properties which are independent of temperature. It is finally assumed that the top layer is not influenced by the rest of the bed e.g. through conduction. With these assumptions, it can be derived from the thermal energy balance on a particle that,

$$\Phi_q = A_{sf}h_{sf}^{a} \left( \langle T \rangle^T - \langle T \rangle^p \right)$$

wherein the interfacial area is taken as the surface of a spherical of diameter $d_{p}$. With a step change in the fluid inlet temperature from $\langle T \rangle^T_0$ to $\langle T \rangle^T_1$ on $t=0$ and with the top of the bed initially at uniform temperature $\langle T \rangle^p_0$, the initial and boundary conditions of the model are

$$\langle T \rangle^T_0 = \langle T \rangle^T_0 \quad t < 0$$

$$\langle T \rangle^T_0 = \langle T \rangle^T_0 \quad t \leq 0$$

$$\langle T \rangle^T_1 = \langle T \rangle^T_1 \quad t \geq 0$$

With these boundary conditions the solution of the model equation 3.7 written in terms of the characteristic time $\tau$ of the heating process is

$$\frac{\langle T \rangle^T_1 - \langle T \rangle^T_0}{\langle T \rangle^T_1 - \langle T \rangle^T_0} = e^{-\frac{t}{\tau}}$$

$$\tau = \frac{d_{p}(\rho c_{p})^s}{6h_{sf}^{a}}$$

Figure 3.8 Schematic representation of the single-phase model
In the above derivation a step-change in the gas inlet temperature was assumed. In the experiments a step change was not possible to attain. As mentioned, the gas inlet temperature did, after the initial step-change, continue to rise linearly with a slope $\beta$. For such conditions the boundary conditions to the model equations become

\[
\begin{align*}
\langle T \rangle' & = \langle T \rangle_0^s \quad t < 0 \\
\langle T \rangle^s & = \langle T \rangle_0^s \quad t \leq 0 \\
\langle T \rangle' & = \langle T \rangle_0^s + \beta \cdot t \quad t \geq 0
\end{align*}
\] (3.13-3.15).

This leads to an exact solution of the transient heating behaviour in which the observed characteristic time is influenced by the raising gas inlet temperature according to,

\[
\langle T \rangle^s = \beta \cdot t + \langle T \rangle_1^f - \beta \cdot \tau + \left( \langle T \rangle_0^s - \langle T \rangle_1^f + \beta \cdot \tau \right) \cdot e^{-\frac{t}{\tau}}
\] (3.16)

\[
\tau = \frac{d_p (\rho c_p)^s}{6h_{sf}}
\] (3.17).

With both solutions, the interfacial transfer coefficient can be determined from the experiments through the characteristic time of the temperature response in the top of the bed. From figure 3.9, which shows both solutions for the same heat transfer coefficient and temperature step, it can be seen what the significant influence will be of neglecting the rise in gas inlet temperature in which the extended model denotes equation 3.16-3.17 and the simple model denotes equation 3.11-3.12. In the simple model, it is necessary to presume an equilibrium temperature which, as would have been done in the analysis, is taken as the final measured temperature in the analysis.

![Figure 3.9, the influence of rising gas temperature](image)

*Heat transport in the packed bed*
3.3.3 Schuhmann model

Even when the temperature response of the top layer can be accurately measured, it is at least doubtful whether the first layer of the bed will give an accurate estimate of the average heat transfer coefficient within the bed. Therefore, it is more accurate to measure the response of the bed at different levels within the bed. However, directly under the first layer of the bed, the fluid temperature is influenced by heat transfer to the solid phase. In order to describe this influence it is needed to set up a thermal energy balance for the fluid phase.

\[
\Phi_q = A_{sf} \cdot h_{sf} \cdot \varepsilon (\langle T \rangle_f - \langle T \rangle_s)
\]

\[ \varepsilon = \frac{V_f}{V_{bed}} \]

In the model, graphically represented in figure 3.10, the fluid phase as well as the solid phase are each assumed to have a uniform temperature at a given depth in the bed \(z\) and the model is thus one dimensional. The fluid phase is further assumed to have a constant velocity \(u_x\) over the entire cross section of the bed. With the thermophysical properties of both fluid and solid phase being independent of temperature, the energy equation of fluid and solid phase are given by

\[
\frac{\partial \langle T \rangle_f}{\partial t} + \langle u_x \rangle_f \frac{\partial \langle T \rangle_f}{\partial z} = \frac{A_{sf}}{\varepsilon V_{bed} (\rho c_p)_f} h_{sf} (\langle T \rangle_s - \langle T \rangle_f)
\]  
(3.18)

\[
\frac{\partial \langle T \rangle_s}{\partial t} = \frac{A_{sf}}{(1-\varepsilon)V_{bed} (\rho c_p)_s} h_{sf} (\langle T \rangle_f - \langle T \rangle_s)
\]  
(3.19).

With a step change in the fluid inlet temperature and with a bed that is initially at uniform temperature the following initial and boundary conditions of the model hold:

\[
\langle T \rangle_f = \langle T \rangle^0_0 \quad t < 0, \forall z
\]  
(3.20)

\[
\langle T \rangle_s = \langle T \rangle^0_0 \quad t \leq 0, \forall z
\]  
(3.21)

\[
\langle T \rangle_f = \langle T \rangle^1_t \quad t \geq 0, z = 0
\]  
(3.22).
With these boundary and initial conditions, the exact solution of equation 3.18 and 3.19 originally derived by Schuhmann \(^1\) can be written as

\[
\frac{\langle T \rangle_0^z - \langle T \rangle_1^z}{\langle T \rangle_0^z - \langle T \rangle_1^z} = e^{-(T+Z)} \sum_{n=1}^{\infty} T^n M_n(TZ) \tag{3.23}
\]

\[
\frac{\langle T \rangle_0^t - \langle T \rangle_1^t}{\langle T \rangle_0^t - \langle T \rangle_1^t} = e^{-(T+Z)} \sum_{n=0}^{\infty} T^n M_n(TZ) \tag{3.24}
\]

\[
Z = \frac{6h_{ef}Z}{\langle u \rangle^2 (1 - e)(\rho c_p)^2 \rho d_p} \quad T = \frac{6h_{ef}t}{(\rho c_p)^2 (1 - e)^2 d_p} \tag{3.25, 3.26}
\]

\[
M_n(TZ) = I_0(n2\sqrt{TZ}) \cdot (TZ)^{n/2} \tag{3.27}
\]

in which \(I_0\) is the modified Bessel function of the first kind, \(Z\) is the dimensionless depth in the bed and \(T\) the dimensionless time. A graphical representation of the solution is more providing than the mathematical equations and is given in figure 3.11.

![Graphical Representation](image)

**Figure 3.11** Graphical representation of the exact solution to the Schuhmann model

At the top of the bed \((Z=0)\), this solution reduces to the equation of the simple model. With respect to the quasi single-phase model, it is significant to note that the difference in response of the top of the bed and a layer at a slightly larger depth can be very substantial. For example, the difference in response between the first thermocouple and the top of the bed, at a bed voidage of 0.41 and a flow of 279 Nm\(^3\)/hr is given by the difference between the curve of \(Z=0\) and \(Z=4\) in figure 3.11. This further indicates the limited applicability and accuracy of the simple quasi single-phase model. In effect, the single-phase model is most likely to give an underestimation of the heat transfer coefficient.

A strong limitation in the use of the solution of Schuhmann model in analysing the pilot scale experiment is the fact that it can not deal with process conditions present in the experiments. The slope in the inlet fluid temperature and an axial non-uniform initial temperature lead to initial and boundary conditions for which no exact solution is available. Numerical solution methods have to be reverted to.
It is clear from figure 3.12 that the internal heat transport can not be neglected in the analysis of the experiments and could even be the dominant resistance depending on the exact value of the interfacial heat transfer coefficient and the effective conductivity.

It is possible to include the internal heat transport phenomena in the Schuhmann and quasi single-phase model by replacing the interfacial transfer coefficient for the apparent overall heat transfer coefficient as given in equation 3.31. The transport term can now be viewed as combining both the resistance for transport from the fluid to the solid boundary and the internal solid conduction resistance:

\[
\rho c_p \frac{\partial T_f}{\partial t} = h_f \left( T_f - T_s \right) - \frac{\partial}{\partial r} \left( \frac{k_s}{r} \frac{\partial T_s}{\partial r} \right) + Q
\]

(3.33)

It has to be emphasised that this is a crude way of modelling the effects of internal resistance, especially when the internal resistance is relative large compared to the external resistance. Furthermore, it is not possible to include effects which depend specifically on the solid surface temperature e.g. axial conduction and radiation in the solid phase. In both cases a model which includes the internal temperature profile will be necessary.

### 3.3.5 Particle-based model

In the single particle model, the solid phase is no longer assumed to be of uniform temperature at each depth in the bed. The internal heat transport is modelled using an effective conduction model which, for a spherical shaped solid phase leads to

\[
\frac{\partial T_s}{\partial t} = \frac{\lambda_{\text{eff}}}{(\rho c_p)^s} \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T_s}{\partial r} \right)
\]

(3.34)

\[- \lambda_{\text{eff}} \frac{\partial T_s}{\partial r} = h_{\text{eff}} \left( T_s \right)_{\text{surface}} - \left( T_f \right) \quad r = \text{solid, } \forall t \forall z
\]

(3.35)

\[- \lambda_{\text{eff}} \frac{\partial T_s}{\partial r} = 0 \quad r = 0, \forall t \forall z
\]

(3.36)

in which \( r \) denotes the radial co-ordinate within the solid phase which is taken as being spherical symmetric. In this model it is assumed that the density of the solid is independent of
temperature. The assumptions on the fluid phase have not changed from the Schuhmann model so the equation describing the fluid phase,

\[
\frac{\partial \langle T \rangle^f}{\partial t} + \langle u_z \rangle^f \frac{\partial \langle T \rangle^f}{\partial z} = \frac{A^{sf} h^{sf}}{d V^{bed}} \left( T_s \right)_{\text{surface}} - \langle T \rangle^f
\]

(3.37)

differs only by the surface temperature of the solid introduced in the driving force of the transfer term. In order to arrive at an exact solution, it was necessary in the Schuhmann model to assume constant thermophysical properties of both solid and fluid phase. However, the model equations do not change if the specific heats are taken as temperature dependent. The same is true for the fluid density which can be verified by the derivation of equation 3.37 from the thermal energy balance and the continuity equation;

\[
\frac{\partial \rho^f}{\partial t} + \frac{\partial}{\partial z} \left( \rho (u_z)^f \right) = 0
\]

(3.38)

\[
c_p^f \frac{\partial}{\partial t} \langle \rho (T) \rangle^f + c_p^f \frac{\partial}{\partial z} \langle \rho (u_z) (T) \rangle^f = \frac{A^{sf}}{d V^{bed}} h^{sf} \left( T_s \right)_{\text{surface}} - \langle T \rangle^f
\]

(3.39)

\[
\left( c_p \rho \right)^f \frac{\partial}{\partial t} \langle T \rangle^f - \langle \rho (u_z) \rangle^f c_p^f \frac{\partial}{\partial z} \langle T \rangle^f = \frac{A^{sf}}{d V^{bed}} h^{sf} \left( T_s \right)_{\text{surface}} - \langle T \rangle^f
\]

(3.40).

The product of density and velocity is constant over the entire bed as it is the fluid mass flux based on the area of the fluid and is assumed constant over the temperature range in the bed. As the equation of the fluid phase is related to a unit of fluid volume and the equation of the solid phase is only for one particle, the expression for the interfacial area is given by:

\[
A^{sf} = V^s \frac{A^s}{V^s} = V^s \frac{6}{d_p}
\]

(3.41)

\[
A^{sf} = (1 - \varepsilon) V^{bed} \frac{6}{d_p}
\]

(3.42)

With a step change in the fluid inlet temperature (either or not followed by a continuous rise), and a bed that is initially at uniform temperature, the following initial and boundary conditions hold:

\[
\langle T \rangle^f = T_0^s \quad t < 0 \ \forall z
\]

(3.43)

\[
T^s = T_0^s \quad t \leq 0, z \leq \frac{d_p}{2} \ \forall z
\]

(3.44)

\[
\langle T \rangle^f = \langle T \rangle^f_{1} + \beta \cdot t \quad t \geq 0, z = 0
\]

(3.45).

For these initial and boundary conditions with $\beta=0$, an exact solution exists by Ivantsov. However, when the conditions of uniform initial temperatures and constant gas inlet temperature do not hold, an exact solution is no longer available. In that case, numerical solutions have to be reverted to. The advantage of numerical schemes is that the model can be easily extended to incorporate different transport mechanisms and (heat) effects, for example the heat effects of the wall. Furthermore, it was found that the inlet gas temperature and the inlet flow fluctuated during the experiment, this could be incorporated in the model as well.
3.3.4 Overall heat transfer coefficient

The transport of heat from the fluid to the solid phase consists of two consecutive processes. As explained before, first heat is to be transported to the solid boundary before it is further transported within the solid phase. However, the Schuhmann and the quasi single-phase models do not include this second process as it is assumed that the internal heat transfer can be neglected. This assumption of infinite conductivity should be checked, especially since the solid conductivity in the experiments is very low, as found in the previous chapter.

As was done in the previous chapter, the two transport processes can be described by resistances in series, related inversely with the heat transfer coefficient. These resistances can be replaced by a single overall resistance $R^{\text{tot}}$

$$R^{\text{tot}} = R^{\text{external}} + R^{\text{internal}}$$  \hspace{1cm} (3.28)

$$\frac{1}{h^{\text{tot}}} = \frac{1}{h^{\text{sf}}} + \frac{1}{h^{\text{cond}}}$$  \hspace{1cm} (3.29)

where $R^{\text{external}}$ and $R^{\text{internal}}$ denote the internal and external resistance respectively, $h^{\text{tot}}$ the apparent overall heat transfer coefficient. The conductive heat transfer coefficient is denoted by $h^{\text{cond}}$ and is for a spherical particle of constant thermophysical properties given by

$$h^{\text{cond}} = \frac{6.6 \lambda^{\text{eff}}}{d_p}$$  \hspace{1cm} (3.30)

for sufficient long times $^{17}$. In this equation $\lambda^{\text{eff}}$ is the effective conductivity of the solid as described in the previous chapter. Including this expression for the conductive heat transfer coefficient in equation 3.27, the dependency of the apparent overall heat transfer coefficient and the interfacial heat transfer coefficient becomes

$$h^{\text{tot}} = \frac{1}{\left(\frac{1}{h^{\text{sf}}} + \frac{6.6 \lambda^{\text{eff}}}{d_p}\right)}$$  \hspace{1cm} (3.31).

Depending on the properties of the phases and the process conditions, the relative importance of internal or external resistance can vary. By comparing the resistances using,

$$\frac{R^{\text{internal}}}{R^{\text{external}}} = \frac{d_p}{6.6 \lambda^{\text{eff}}} = \frac{d_p h^{\text{sf}}}{\lambda^{\text{eff}}} \frac{1}{6.6} = \frac{Bi}{6.6}$$  \hspace{1cm} (3.32)

it is possible to determine which process is dominant. For the expected range of interfacial heat transfer coefficients (50-500 W/m²K) and effective solid conductivities (0.3 - 1.2 W/mK) this analysis is given in figure 3.12.
3.3.6 Wall effects and heat losses

An important complication in modelling the packed bed used in the experiments are, as mentioned, the effects induced by the finite size of the bed. In short, this will mean that there is no uniformity in temperature and flow over a cross section of the bed which has been assumed in all aforementioned models. This is caused by the fact that heat is transported from both phases to the wall. When this is very low due to good insulation, it can be neglected but in the experiments it turned out to be very significant as was shown in the experimental section.

In order to deal with the heat losses, a one dimensional model of the bed will no longer be sufficient as radial heat transport will have to be included leading to a much more complicated mathematical model and eventually to a model like equation 3.4-3.5. In this project it has been attempted to incorporate heat losses in the one dimensional model as the measurements have only been at the centre of the bed.

3.3.6.1 Centre of the pot approach

In a first approach, as was used in the quasi single-phase model, it is assumed that in the top part of the bed the heat losses have not yet had a significant effect on the thermal balance over the centre section of the bed. This can be verified by assuming that the heat losses to the wall have to penetrate towards the centre in an effective conduction process. The effective radial conductivity of the bed is estimated at 100 times better than the gas conductivity $^{10}$. With this estimation, it can be estimated to where the wall effects have penetrated by using the theory of the penetration depth, in which the penetration depth indicates the position where a 20% or less change in temperature has occurred relative to the total driving force $^{20}$. The results are given in figure 3.13 for a selection of conditions used in the experiments from which the range of the effect can be estimated. The full analysis method is given in appendix F.

![Graph](image_url)

Figure 3.13, estimated penetration depth in time, relative to the centre for various conditions which are given in the legend; flow rate, bed voidage and average temperature.

With an average experiment lasting approximately 1000s it can be seen that it is very likely that the temperature at the centre of the bed is already influenced by the presence of the heat losses to the wall. This effect will be worse deeper in the bed, as it is influenced by the heat losses in the part of the bed above it. However, as a first approximation it could be used for the higher regions of the bed.

It can thus be assumed that a centre section of the bed can be modelled as not being influenced by the wall. However, the temperature profile will have an influence on the fluid flow through the centre of the bed as the properties of the fluid influencing the flow (viz.
density and viscosity) are strongly temperature dependent. These effects can be analysed using Ergun's relationship for pressure drop \(^2\)

\[
\frac{\Delta p}{\Delta z} = \frac{150 (1-\varepsilon)^2}{\varepsilon^3} \frac{\eta_f}{\rho_f} \left( \Phi_m^f \right) \frac{1.75 (1-\varepsilon)}{\varepsilon^2} \frac{1}{\rho_f} \left( \frac{\Phi_m^f}{A_{bed}} \right)^2
\]

in which \(\Delta p/\Delta z\) denotes the pressure drop per unit of bed height, \(\Phi_m^f\) the fluid mass flow, \(A_{bed}\) the cross sectional area of the bed and \(\eta_f\) the dynamic viscosity of the fluid. To calculate the flow difference resulting from the temperature difference between the inner section area \((A_1)\) and the outer section area \((A_2)\), as shown in figure 3.14, it is assumed that both sections have a uniform temperature and the same pressure drop.

![Figure 3.14, sections of the pot used in flow calculations](image)

It is assumed that the inner section of the bed has a temperature which is 50 K larger than the outer section. The validity of this assumption can be checked by comparing the radial conduction through the bed, as used earlier with the actual heat loss of about 20K. In the analysis, given in figure 3.14 it is assumed that the temperature within a section with area \(A_1\) or \(A_2\) of the bed can be considered constant in the axial direction.

![Figure 3.15, determination of the difference in centre and wall temperature (cross section across the length of the cylindrical bed)](image)

With an air conductivity of approximately 0.04 W/mK, a fluid temperature difference of 20K and a flow of approximately 0.1 kg/s (300 Nm/hr), this results in a conductive heat transfer...
within the bed of 70 W/m²K and a temperature difference in radial temperature difference of 50 K.

In a first approximation, both sections are assumed of equal area resulting in a radius of \( A_1 \) of 0.12 m. The flow \( \xi \) per unit bed area in the centre section relative to the total flow per unit of bed area can then be calculated by

\[
\frac{\Phi_{m,A_1}}{A_1} = \frac{\Phi_{m,A_1}}{\Phi_{m,A_1} + \Phi_{m,A_2}} \frac{A_1}{A_1 + A_2}
\]

(3.47)

in \( \Phi_{m,A_1} \) and \( \Phi_{m,A_2} \) denote the fluid mass flow of through section 1 and 2 respectively. The dependency of \( \xi \) on flow, temperature and voidage respectively, is given in figure 3.16 according to the calculations of appendix G.

Figure 3.16: percentage of the average flux going through \( A_1 \),

\( \xi = f(\text{flow}, \text{radius of } A_1, \text{bed voidage}, \Delta T(A_1,A_2) \)

0.25 kg/s = 700 Nm³/hr, 0.05 kg/s = 150 Nm³/hr

It can be seen from figure 3.16 that the difference in flux will be very low (< 2%) although the influence will become larger with a smaller area being assumed for \( A_1 \), is taken. However, at high temperatures where the penetration depth will be highest, the flow difference will be the lowest. Therefore, the effect of the heat losses on the flow will not be introduced in further modelling.

3.3.6.2 Model of heat losses to the wall
Another approach to incorporating the heat losses without losing the one dimensional character of the model, is to assume that the heat losses are only drawn from the fluid phase and that the fluid phase in ideally mixed at each height in the bed. In this model the heat transport to the wall has to be quantified. The heat transport to the wall can be divided into convective heat transport from the fluid phase to the surface of the wall and transport within the wall.
The heat transport within the wall is, as in the solid phase, effectively by heat conduction. As the wall heats up only slowly, the heating of the wall will be a penetration process which is not expected to be effectively modelled in the form of Newton's law of cooling. Therefore, the heat transport within the wall is modelled in detail. It is assumed that in the wall heat flows only in the radial direction and not in the axial as the temperature differences in the radial direction (500-1100 K) are far bigger than those in the axial direction (max. 50K). With the further assumption of constant thermophysical properties and a cylindrical wall geometry the heat transport within the wall \((w)\) is given by

\[
\frac{\partial T^w}{\partial t} = \frac{\Lambda^w}{(\rho C_p)^w} \frac{1}{w} \frac{\partial}{\partial w} \left( \frac{\partial T^w}{\partial w} \right) \tag{3.48}
\]

in which \(w\) denotes the radial position within the wall. This model equation has two boundary conditions. For the boundary condition at the inside of the wall which is in contact with the packed bed, it is assumed that the heat is transferred through convective transport from the fluid phase resulting in

\[
-\Lambda^w \frac{\partial T^w}{\partial w} = h^{wf}(T^w - \langle T \rangle^f) \quad \forall w = \Omega^{inner}, \forall t \forall z \tag{3.49}
\]

wherein \(\Omega^{inner}\) is the inner radius of the wall and \(h^{wf}\) is the interfacial heat transfer coefficient between wall and fluid. For the other boundary, in contact with the environment \((e)\) it is assumed that heat is transferred by (free) convection which leads to

\[
-\Lambda^w \frac{\partial T^w}{\partial w} = h^{we}(T - \langle T \rangle^e) \quad \forall w = \Omega^{outer}, \forall t \forall z \tag{3.50}
\]

with \(\Omega^{outer}\) being the outer radius of the wall, \(h^{we}\) the interfacial heat transfer coefficient between wall and environment and \(\langle T \rangle^e\) being the temperature of the environment. This boundary condition is given to be comprehensive but in the practice of the experiments the heat transport to the environment has very little influence on the heat transport within the wall.

As the heat loss is drawn from the fluid phase, the model of the fluid phase has to be adapted in order to include the heat transport to the wall. This is done by including an extra transport term similar to the term describing the heat flow to the pellets. This results in

\[
\frac{\partial \langle T \rangle^f}{\partial t} + \langle \Omega_z \rangle^f \cdot \frac{\partial \langle T \rangle^f}{\partial z} = \frac{1}{\delta \nu^{bed} (\rho C_p)^f} \left( A^{wf} h^{wf} (T^w_{surface} - \langle T \rangle^f) + A^{wf} h^{wf} (T^w_{surface} - \langle T \rangle^f) \right) \tag{3.51}
\]

in which \(A^{wf}\) this the wall area per unit volume of fluid. This area is related to the dimension and condition of the wall by

\[
A^{wf} = 2A^{inner} \Omega^{inner} dz \tag{3.52}
\]

\[
dz = \frac{\nu^{bed}}{A^{bed}} = \frac{\nu^{bed}}{A^{bed} = \frac{\nu^{bed}}{\pi (\Omega^{inner})^2}} \tag{3.53}
\]

\[
A^{wf} = \frac{2\nu^{bed}}{\Omega^{inner}} \tag{3.54}
\]
3.3.7 Incorporation of the fines

Another complication in the modelling of the packed bed is how to incorporate the influence of the fines. The influence on the bed voidage can easily be incorporated as it can be calculated from directly measurable quantities like resultant bed height and mass of the solid material. The other influences on the model are not straightforward and include interfacial area and influence on the flow pattern. The flow pattern is not used in the above mentioned models and will have to be lumped into the heat transfer coefficient. The fines are thought to have a large influence on the flow, and thus on the heat transfer, as they can effectively block the pores in the structure of the larger pellets.

In the above models the interfacial area as well as the volume of a particle has been used to estimate the heating behaviour of the bed. For pellets, which can very well be assumed spherical and uniform in size, no problem is encountered. For the fines this is totally different. They do not have a uniform size or shape. The only known characteristics of the fines are their total weight and their apparent size from the sieving fraction. An assumption on the shape of the fines will be necessary in order to estimate the total area of the fines. The assumption of a spherical shape will probably give a good lower estimate. If necessary this can be refined using a shape factor.

In another option, the interfacial area of the fines is not calculated as the error in this calculation is assumed to be to great not only by the fact that the shape is not known, but also by the fact that not all of the available interfacial area might be used in the transfer of heat. In this approach the interfacial area is not used in the calculation of the heat transfer coefficient but instead the total volume of the bed is used, resulting in an overall volumetric heat transfer coefficient $h'_{vot}$ \(=\) \(\frac{A_{tot} h_{tot}}{V_{bed}}\) \(\text{(3.55)}\), where $A_{tot}$ is the total interfacial area calculated using the assumption of spherical fines and $V_{bed}$ is the volume of the bed. In fact, the total heat transfer coefficient can be regarded as given the amount of heat transferred to a unit of the bed due to a temperature difference of 1 degree between the packed bed and the gas.

The next issue is on how to incorporate the fines in the heat transfer equations. Naturally they will have a different response than the bigger pellets. A number of options exist for this situation. In this project the following option has been used as it was used in the existing model of the pellet induration process (BRAM, see general introduction). This would ensure that the effects of lumping different effects would not be different. In this option, the fines are modelled as a number of extra pellets whose weight equal the total weight of the fines. In order to compare the results of experiments of different bed voidage, the overall volumetric heat transfer coefficient has been used.

As was mentioned in the introduction, one of the incentives of the investigation has been the expected underestimation of the influence of fines on the heat transfer in a packed bed. The way in which the are modelled in BRAM will not be physically realistic, as the average temperature of the small fines is likely to rise much quicker than the temperature of the larger pellets. Generally, the effect of fines or more specifically, the influence of multi-granular packed beds, is modelled using a volume averaged diameter. The average diameter $\langle d \rangle$ of the particles in the packed bed is given by

$$\langle d \rangle = \frac{1}{\left( \frac{V_1}{V_{tot} d_1} + \frac{V_2}{V_{tot} d_2} + \cdots \right)}$$ \(\text{(3.56)}\).
in which $V_i$ denote the volume of the fraction particles with diameter $d_i$ etc. and $V_{tot}$ is the total volume of the particles. In this approach has much of the same disadvantages as the above mentioned method, although the influence of the faster heating of fines has been adapted for a little. It is probably a very good estimation when the particles differ only slightly in size. With a very distinct difference in size, as is the case in the additions of fines to the pellet bed, two mechanisms of very different time scale are lumped which is not expected to give an accurate description. The following description is thought to be improvement.

As the fines are significantly smaller size, they will heat up much faster than the pellets showing the approximate same behaviour as the pellet outer boundary. Because of their size, the fines will also have a much larger interfacial area and will be in closer contact with the pellet than the neighbouring pellets. Due to their higher temperature, high interfacial area and improved contact with the pellets, the fines could be thought of as conducting heat from the gas to the pellet. In such a model they can be regarded as 'heating fins' of the pellet. This influences combined with the expected influence of the fines on the flow in the bed were reason to conduct tests on the pilot scale set-up.
3.4 Numerical implementation

For both the quasi single-phase and the standard Schuhmann model, an exact solution can be found for the temperature response of the bed. For the Schuhmann model with a non uniform initial bed temperature and a rising temperature of the gas inlet temperature as well as for the particle based model, no exact solution is available for the partial differential equations (pde's) of the model and numerical solution methods have to be reverted to.

3.4.1 Discretisation method

As in the previous chapter, the pde's of the model are numerically solved in two steps. First, the pde is made discrete in the space co-ordinate, before the resultant ordinary differential equation (ode) is numerically integrated over the required time span. In the previous chapter, the model consisted of one pde whereas the model of the pot consists of a system of either two or three (including the wall) partial differential equations. This poses no principle difference, as each of these equations is one dimensional and only interacts with the variables of the other pde's through the boundary conditions. In this case each of the pde's can be made discrete in their own dimensional co-ordinate.

As explained in the previous chapter, the first step in the discretisation of a pde is the construction of a grid which consists of a finite number of points at which the value of the dependent variable, in this case either the solid, the fluid or the wall temperature, will be determined. As in the previous chapter the control volume method\textsuperscript{23} is used to discretise each of the pde's in which the volume of each of the phases is divided into a number of non-overlapping control volumes, such that there is one control volume surrounding each grid point. The grid points are located in the centre of the control volume.

The pde can now be regarded as originating from a thermal energy balance over this control volume. Next, the pde is integrated over each control volume. The terms in the differential equations which are differentials over the space co-ordinate, i.e. the conduction terms in the solid and wall and the convective term in the phase, are evaluated using piece-wise continuous profiles. These profiles express the variation of the dependent variable between the grid points.

Using the control volume method, the pde model equations can be transformed into a finite number of ode's in the temperatures at those grid points.

3.4.2 Discretisation of the packed bed

As was stated above, the bed is to be divided into control volumes in order to numerically solve the model equations. The way in which the control volumes are located in the packed bed (including the wall) is given schematically in figure 3.17 and will be clarified below.

![Figure 3.17, co-ordinate system in the numerical implementation of the packed bed](image-url)
3.4.2.1 Fluid phase

First of all the bed is divided in the axial direction into Z volumes of cylindrical shape and of thickness \( \delta z \). Such a volume of the bed can be divided into a volume of solid and a volume of fluid using the bed voidage. The model equation of the fluid phase can directly be applied to this fluid volume. All the control volumes are of equal size and the grid point is located in the centre of the control volume as shown in figure 3.18.

**Figure 3.18, summary of control volume method for the fluid phase**

With the nomenclature of figure 3.18, the discretisation of equation 3.18 or 3.37 for the fluid phase this leads to

\[
\int_{w}^{e} \left( \rho c_p \frac{\partial}{\partial t} (T)^f \right) dz + \int_{w}^{e} \left( F \rho c_p \frac{\partial}{\partial z} (T)^f \right) dz = \int_{w}^{e} \left( S_c + S_p (c_p (T))^f \right) dz
\]

\[ (\rho c_p) \frac{d}{dt} \left( (T)^f \right)_{w} + F \left( (c_p (T))^f - (c_p (T))^f \right) = \left( S_c + S_p (c_p (T))^f \right) \Delta z \]  

(3.57)  

(3.58)

in which \( F \) denotes the mass flow of fluid per unit of cross sectional area of the fluid flow \( = \rho'(u^f)' \). Furthermore, according to Patankar the source term is not specified but divided into a constant part \( S_c \) and a part \( S_p \) with a linear dependency on the volumetric enthalpy which is evaluated at the temperature of the grid point. The source term has not been specified as it is dependent on the model used.

In order to further discretise equation 3.58 it is necessary to introduce a piece-wise continuous profile for the temperature (or for the internal heat when the specific heat is taken as temperature dependent) over the space co-ordinate \( z \). A piece-wise linear profile as used in the previous chapter, is not suitable for the fluid phase as it would lead to instability. A well-known solution to this problem is the use of the 'upwind scheme'. In the 'upwind scheme' the value of the heat at an interface is equal to the value of the heat at the grid point on the upwind side (opposite of the flow direction) of the interface.

\[
(c_p (T))^f_w = (c_p (T))^f_p
\]

\[
(c_p (T))^f_e = (c_p (T))^f_p
\]

(3.59)  

(3.60)

This can physically be regarded as each control volume being a ideally mixed reactor. Other more elaborate piece-wise continuous profiles are possible which are more suitable to including hydrodynamic dispersion in the fluid phase. However, for a purely convective model as is used here, the upwind scheme is best suited. Using equation 3.59 and 3.60 in equation 3.58 a complete discretisation of the fluid phase model can be attained:

\[
\frac{d(T)^f_p}{dt} = \frac{1}{(\rho c_p) \Delta z} \left[ F \left( (c_p (T))^f - (c_p (T))^f \right) - \left( S_c + S_p (c_p (T))^f \right) \Delta z \right]
\]

(3.61)
This leaves the source term to be specified. These source terms consist of the interfacial heat transport terms to the solid phase and depending on the model also to the wall phase. From the model equations it can be derived that for the particle based model

\[ S_C = \frac{6h_f \epsilon (T^s_{\text{surface}} - T)}{d_p \cdot \epsilon} \] (3.62)

\[ S_P = \left( \frac{6h_f (1 - \epsilon)}{d_p \cdot \epsilon} \right) \frac{1}{c_p} \] (3.63)

in which the solid surface temperature has to be replaced with the average solid temperature when the Schuhmann model is used. When the heat losses to the wall are modelled as well the source term is given an additional term for transport to the wall

\[ S_C = \frac{6h_f (1 - \epsilon) T_s^{\text{surface}}}{d_p \cdot \epsilon} + \frac{2h_f}{\Omega_{\text{inner}} \epsilon} T_W^{\text{surface}} \] (3.64)

\[ S_P = \left( \frac{6h_f (1 - \epsilon)}{d_p \cdot \epsilon} + \frac{2h_f}{\Omega_{\text{inner}} \epsilon} \right) \frac{1}{c_p} \] (3.65)

The entire discretisation of the fluid model equation will result in Z ode’s in the temperatures at the Z grid points. In the numerical model, the measured inlet gas flow rate and gas inlet temperature at time in the experiment have been used as boundary conditions.

3.4.2.2 Solid phase

In each layer of the bed the volume of the solid phase will consist of a large number of particles. Within one layer of the bed size \( \delta z \), these particles are all assumed to have the same behaviour, so the solid phase can be modelled by looking at one particle.

In case of the Schuhmann model, the solid phase is assumed to be of uniform temperature. In this case, the governing equation 3.18 can be directly applied to the volume of one particle resulting in

\[ d \left( T^s \right) \frac{d t}{d} = \frac{6h_f}{d_p (\rho \cdot c_p)} \left( \langle T \rangle^f - \langle T \rangle^s \right) \] (3.66)

for each layer of the bed. In total this would add Z ode’s to the description of the packed bed. When the particle based model is used, a grid within the particle is necessary. In this case, as was done in the previous chapter, the (spherical) particle is divided into N-2 concentric spherical volumes of thickness \( \delta r \) and 2 volumes of thickness \( \delta r/2 \) to accommodate the boundary conditions as shown in figure 3.19.

![Figure 3.19, summary of the control volume method for the solid phase](image)

('C' denotes the pellet centre, 'B' denotes the surface boundary)
Numerical implementation

The model equation 3.34 of the solid phase is next made discrete using this grid. For all the non-boundary control volumes this leads to

\[
\frac{dT_p^s}{dt} = \frac{1}{3 \left( r_0^3 - r_w^3 \right) \rho c_p} \left( r_p^2 \lambda_{p}^{\text{eff}} \frac{\partial T_p^s}{\partial r} \bigg|_{r_p} \right) - \frac{r_w^2 \lambda_{w}^{\text{eff}} \frac{\partial T_w^s}{\partial r} \bigg|_{r_w}}{3 \left( r_0^3 - r_w^3 \right) \rho c_p}
\]

which is the same as in the previous chapter. However, in this case the conductivity is not assumed independent of the temperature. As the temperature and thus the conductivity is only known at the grid points, the calculation of the conductivity at the interfaces of the control volumes is not straightforward. In order to prevent accumulation in the interfaces, the heat flux calculated from either side of the interface should be the same. It can be shown that this is achieved when the conductivity at the interface is taken as the harmonic mean of the conductivities at grid points on either side of the interface given by

\[
\lambda_{I}^g = \frac{2 \lambda_{E}^{\text{eff}} \lambda_{P}^{\text{eff}}}{\lambda_{E}^{\text{eff}} + \lambda_{P}^{\text{eff}}}
\]

(3.68)

\[
\lambda_{I}^w = \frac{2 \lambda_{W}^{\text{eff}} \lambda_{P}^{\text{eff}}}{\lambda_{W}^{\text{eff}} + \lambda_{P}^{\text{eff}}}
\]

(3.69)

This approach is also valid when the conductivity is dependent on position, as with the transition from one material to another in the wall. At the solid outer surface, the convective boundary condition of 3.35 in discretised form is given by

\[
\frac{dT_B^s}{dt} = \frac{1}{3 \left( r_i^3 - r_B^3 \right) \rho c_p} \left( r_i^2 \lambda_i^{\text{eff}} \frac{\partial T_i^s}{\partial r} \bigg|_{r_i} - \frac{r_B^2 h_{\text{eff}} \left( T_B^s - T_i^s \right) f}{3 \left( r_i^3 - r_B^3 \right) \rho c_p} \right)
\]

(3.70)

in which the control volume is half the size of the central control volumes and in which the grid point is located on the outer boundary of the control volume, but on the outer boundary. The same is true for the control volume at the centre of the spherical particle where a zero flux through the centre is assumed.

\[
\frac{dT_C^s}{dt} = \frac{1}{3 \left( r_i^3 \rho c_p \right)^{\frac{1}{3}}} \left( r_i^{2 \lambda_i^{\text{eff}}} \frac{\partial T_i^s}{\partial r} \bigg|_{r_i} \right)
\]

(3.71)

The total discretisation of the solid phase for the particle based would give N*Z ode’s describing the behaviour of the solid phase leading to Z*(N+1) equations for the entire bed.

3.4.2.3 Wall phase

When the heat losses to the wall are modelled as well, a grid is necessary within the volume of the wall associated with a layer in the bed size δz. In the numerical implementation the model for the wall is very similar to the model of the solid phase. Both are effectively conduction processes the only difference being the geometry which is cylindrical instead of spherical. The grid of the wall is build up from M-2 concentric cylindrical volumes of size δφ and 2 volumes of size δφ/2 as shown in figure 3.20.
Numerical implementation

Figure 3.20, summary of the control volume method for the wall phase
('Bo' denotes the outer, and 'Bi' the inner surface boundary)

With the nomenclature of figure 3.20 the discretisation of equation 3.48 for the central control volumes leads to

\[
\frac{(\rho c_p)^w}{\omega} \frac{\partial T^w}{\partial t} = \frac{\partial}{\partial \omega} \left( \omega \lambda^w \frac{\partial T^w}{\partial \omega} \right)
\]

\[(3.72)\]

\[
\frac{(\rho c_p)^w}{\omega} \int_0^\omega \frac{\partial T^w}{\partial t} d\omega = \frac{\partial}{\partial \omega} \left( \omega \lambda^w \frac{\partial T^w}{\partial t} \right) d\omega
\]

\[(3.73)\]

\[
\frac{1}{2} \left( \omega_i^2 - \omega_f^2 \right) \left( \rho c_p \right)^w \frac{dT^w_{Bo}}{dt} = \frac{\omega_i \lambda_i^w (T^w_{\text{Bo}} - T^w_i)}{\delta \omega} - \frac{\omega_f \lambda_f^w (T^w_{\text{Bo}} - T^w_i)}{\delta \omega}
\]

\[(3.74)\]

\[
\frac{d T^w}{dt} = \frac{1}{2} \left( \omega_i^2 - \omega_f^2 \right) \left( \rho c_p \right)^w \left\{ \frac{\omega_i \lambda_i^w (T^w_{\text{Bo}} - T^w_i)}{\delta \omega} - \frac{\omega_f \lambda_f^w (T^w_{\text{Bo}} - T^w_i)}{\delta \omega} \right\}
\]

\[(3.75)\]

wherein it should be noted that the conductivities at the interfaces are calculated according to equation 3.68 and 3.69, to include the transitions between different layers of material within the wall. The discretisation of the inner and outer surface boundary condition are again very similar to those of the solid phase. For the outer surface in contact with the environment this results in

\[
\frac{(\rho c_p)^w}{\omega} \frac{d T^w_{Bo}}{dt} = \frac{1}{d \omega} \left( \omega (-\lambda)^w \frac{d T^w}{d \omega} \bigg|_{\omega=\omega} - \Phi_{\text{q,convection}} \right)
\]

\[(3.76)-(3.77)\]

\[
\frac{d T^w_{Bo}}{dt} = \frac{1}{2} \left( \omega_i^2 - \omega_f^2 \right) \left( \rho c_p \right)^w \left\{ \frac{\omega_i \lambda_i^w (T^w_{\text{Bo}} - T^w_i)}{\delta \omega} - \frac{\omega_f \lambda_f^w (T^w_{\text{Bo}} - T^w_i)}{\delta \omega} \right\}
\]

\[(3.78)-(3.79)\]

and for the wall inner surface in contact with the packed bed to

\[
\frac{(\rho c_p)^w}{\omega} \frac{d T^w_{Bi}}{dt} = \frac{\Phi_{\text{q,convection}}}{2\pi} - \omega (-\lambda)^w \frac{d T^w}{d \omega} \bigg|_{\omega=\omega}
\]

\[(3.80)-(3.81)\]

\[
\frac{d T^w_{Bi}}{dt} = \frac{1}{2} \left( \omega_i^2 - \omega_f^2 \right) \left( \rho c_p \right)^w \left\{ \frac{\omega_i \lambda_i^w (T^w_{\text{Bi}} - T^w_i)}{\delta \omega} - \frac{\omega_f \lambda_f^w (T^w_{\text{Bi}} - T^w_i)}{\delta \omega} \right\}
\]

\[(3.82)-(3.83)\]

In total the description of the wall adds another \(Z^*M\) ode's to the description of the packed bed.
3.4.3 Integration scheme and fit procedure

After the discretisation of the model equations describing the behaviour of the bed, the model of the packed bed is transformed to a set of a number of ode's which is dependent on the model with a maximum of Z(N+1+M) equations.

These equations can be solved using a Gear-type algorithm\textsuperscript{24,25} for which for more information is referred to the previous chapter. However, in the model including the wall, there remain three parameters which have to be determined viz. the different heat transfer coefficient $h^w$, $h^{ww}$ and $h^{wf}$. As was mentioned before the heat transfer coefficient for wall to the environment $h^{ww}$ is not very important in the experiments as the main heat losses are due to the heating of the wall itself which is still in a heat penetration process within the time scale of the experiment. Remain the fluid to wall heat transfer coefficient and the interfacial heat transfer coefficient (from fluid to solid phase). The former of these can be fitted to the quasi steady state temperature gradient in the bed. In that situation, almost all of the heat is transferred to the wall. Using this technique the value of the heat transfer coefficient is found to be approximately 60 W/m$^2$K which resembles remarkably good the estimation of the same heat transfer coefficient determined in the theory section $h^{wf}$.

The interfacial heat transfer coefficient is now the only undetermined parameter in the model and can be determined from the experimental data by a similar $\chi^2$ minimisation method\textsuperscript{26,27} as was used in the experiments on the internal heat transfer in the previous chapter.
3.5 Results and discussion

3.5.1 Experimental difficulties

During the experiments and in the analysis of the experimental data, several problems have been encountered, some of which have been mentioned before, e.g. heat losses. These problems will be focused upon first, as they have had a large influence on the modelling necessary to analyse the experiments. In general, it can be said that the problems mentioned below have made the determination of the heat transfer coefficient difficult and subject to a large error.

3.5.1.1 Heat losses

The first of these problems, which has been mentioned before, has been the loss of heat to the wall of the experimental set-up. Heat losses have occurred in the packed bed as well as in the section above the packed bed where the burner section is located.

The heat losses in the burner section caused the continuous rise in the gas inlet temperature which has been around 0.5 °C/min. As an effect of this, no step change could be attained in which the temperature was constant before and after the step. The main problem with this has been that nor at the beginning of an experiment, nor at the end, the bed has been in a steady state. Due to this, there will always be an axial temperature profile, the effect of which will have to be added to the effects of the heat losses in the packed bed itself which will be discussed next. The modelling of this increase in gas inlet temperature has not been problematic, but it did lead to difficulties as even for simpler models of the bed, it meant that an exact solution was no longer available.

The heat losses to wall within the packed bed have been mentioned a few times already, but at first it was believed that they were very small due to good insulation. Inspection of the experimental data, together with results found in other research conducted with the experimental set-up has changed this image totally. Presently, these heat losses are taken very seriously and have been of grave influence on the results obtainable from the experimental data. The main cause of these heat losses are not so much the loss of heat to the surroundings, but are mainly confined to the gradual heating up of the material of the wall itself and do not reach a steady state in which a constant temperature profile in the wall is attained.

As the heat losses were considered negligible at the time of the experiments, only measurements have been taken at the central axis of the pot. As a result the heat losses could not be determined accurately. An estimate of the heat losses can be obtained from the experimental curves though, as was indicated in the experimental section. After a sufficiently long period following a step-change the bed can be considered quasi steady when the effects of the rise in gas inlet temperature are ignored. In this quasi steady situation the axial temperature gradient can be thought as to be totally due to the heat losses. When in addition to this it is assumed that the gas temperature entering and leaving the bed is ideally mixed in the radial direction, the heat losses can be estimated from their difference in temperature. For the experiments this temperature difference was ± 20 °C which is about 40% of the heat effect induced by the step-change.

A more accurate evaluation of the heat losses can be attained by regarding a heat balance for the bed using experimental data on the inlet and outlet gas temperatures and flows as well as on the temperatures measured at various depth in the bed. For the calculations below, the standard thermocouple measurements have been used.

The heat which has been transferred from the gas while going through the bed is estimated for each interval by subtracting the heat of the gas outlet flow from the heat of the gas inlet flow which have been calculated using an temperature averaged specific heat. The inlet gas flow has been corrected for the gas flow taken up by the suction pyrometer.
The heat taken up by the pellet has been estimated by dividing the packed bed into six sections in the middle of which is positioned one of the standard thermocouples. The temperature of the standard thermocouples, correct for a systematic error which will be discussed in the next paragraph, has been assumed to be an estimate of the bed temperature of the entire section of the bed. The heat of the section has been determined from the temperature by using a specific heat from literature. The result of such an analysis is given in figure 3.21.

Figure 3.21, analysis of the heat transferred from the gas and to the pellet in which the heat content is set to zero at the beginning of each new experiment.

The overall heat losses in each experiment can be determined by comparison of the cumulative heat taken up by the packed bed and the cumulative heat transferred from the gas at the end of the experiment. In the experiments in figure 3.21 this amounts an overall heat loss of around 70%. However, this is highly dependent on the duration of the experiment as after the initial response of the bed, a quasi steady state is observed in which the packed bed barely heats up further. It is therefore better to look at the heat losses occurring at each moment in time. These actual heat losses are obtained by comparing the tangent to the curve of the cumulative heat taken up by the packed bed, and the tangent of the cumulative heat transferred from the gas. These tangents denote the power transferred from the gas and the power taken up by the pellets respectively. The relative amount of power taken up by the packed bed will decrease during the experiments. For example, over the first 250 seconds of the first experiment (figure 3.21, 0-1400s), the average power transferred from the gas amounts to 10 W, while only an average of 5.4 W is taken up by the packed bed. This means that the wall has taken up 4.6 W on average, which is 45% of the total power transferred from the gas. In the quasi steady state of the first experiment, (figure 3.21, 750-1400s) the power transferred from the gas is 4.8 W on average, while the packed bed takes up only 0.4 W which means that the heat losses have risen to 92% of the total power transferred from the gas.

The analysis here is only a rough estimate as the heat content of the bed has been estimated by interpolation between the thermocouple measurements, radial flow differences have been neglected and the gas temperatures have been taken as the average gas temperatures. However, it can be concluded that an very significant part in the order of the heat transferred from the gas, will not be transferred to the gas, but to the wall.

Heat transport in the packed bed
In principle these heat losses do not have to be a problem in the determination of the heat transfer coefficient when they can be modelled sufficiently accurate in a two dimensional model. The problem with this is that there have been no measurements taken in experiments on the radial temperature profile, making the validation of a two dimensional model impossible. Furthermore, the correct inclusion of the heat losses is further hampered by the fact that heat losses in the packed bed are combined with the increase in gas inlet temperature, the dynamic result of which, is difficult to predict.

3.5.1.2 Temperature measurement

In the temperature measurement there have been two kinds of problems; one associated with the logging of the measurements, the other with the actual temperature measured.

As explained in the experimental section, the temperature measurements can be divided into two groups by looking at the apparatus with which they were logged. The standard thermocouples, as well as the inlet and outlet gas temperatures have been measured by the process control system. Additional temperature measurements, as the temperature within a pellet at different heights in the bed, have been logged by a separate system using a personal computer. As the measurements of the temperature inside a pellet are more accurate in the determination of the heat transfer coefficient, the measurements logged by the different two systems have to be combined.

Difficulties have arisen in combining these two measurements. It proved impossible to exactly synchronise the logging of both sources during the time of the experiments but it was thought that the two measurements could be synchronised afterwards, by focusing on a characteristic event in the experiment e.g. the event of shutting down the gas flow at the end of a series of experiments. The temperature measurements of an entire series of subsequent step changes which were logged continuously, one after the other, could thus be synchronised in one step (an example of such a measurement is given in figure 3.22 and 3.23 in which each step change is considered a separate experiment). The actual synchronisation would be achieved by shifting the two sets of logged curves along the time axis in such a way that the chosen characteristic event would coincide.

This failed as by synchronising one characteristic event would cause another characteristic event not to coincide. A growing discrepancy between the two sources was observed when a characteristic event at the beginning of a series of experiments was chosen for the synchronisation. The absolute magnitude of the discrepancy was not very large (1%) but as the different experiments were conducted in series, this could mean a 100s discrepancy in the final experiment. As the logging of the personal computer is very accurate, the conclusion drawn has been that the logging of the process control system is slightly erroneous. This conclusion is supported by data from an earlier investigation on the temperature logging of the experimental set up, which show a deviation of approximately 2% between the time indicated by the process control system and the time measured by stopwatch. The criticality of this error lies in the fact that the discrepancy is not linear and can thus not be corrected for. The source of this problem is thought to be in the fact that in the logging by the process control system, the time needed to change control set-points is not included in the time measurement.

The result of this error is, that there is a uncertainty of around 20s in the starting time of measurement by the personal computer relative to the measurement of process control system.

The second problem with the temperature measurement lies in the actual temperature measured by the different thermocouples. There is a distinct difference between the absolute temperatures measured by the standard thermocouples and the additional thermocouples at approximately the same position in the bed, as can be seen by comparing figure 3.22 and figure 3.23. Both figures contain data from the same series of experiments, figure 3.21 being the measurements of the standard thermocouples and figure 3.23 being the additional
Results and discussion

thermocouple measurements. In order to facilitate the comparison, the inlet and outlet gas temperatures have been plotted in both figures.

![Figure 3.22, example of standard thermocouple measurements](image1)

The temperatures measured by the additional thermocouples are in general significantly lower than those measured by the standard thermocouples during the same experiment at the same position in the bed especially at the higher positioned in the bed.

The fact that one of the additional thermocouples in the top of the bed (11cm) measures a lower temperature than the outlet gas temperature indicates that something is amiss. The use of the correct references and automatic calibration method has been verified so this could not cause the observed difference shown in figure 3.24, which is a copy of 3.23 with the curves of interest being highlighted.

![Figure 3.23, additional thermocouple measurements for the experiment show in figure 3.22](image2)

Heat transport in the packed bed
Results and discussion

Figure 3.24, comparison of gas outlet temperature and additional thermocouple at 11 cm

Furthermore, the effect of the insulation used within the bed leading to a different thermal equilibrium around the thermocouple could be ruled out, since the additional thermocouple at 6 cm is insulated in the same manner also shows a significantly lower temperature as well. This is shown in figure 3.25.

Figure 3.25, comparison of standard thermocouple and additional thermocouple both insulated by a closed ceramic tube. (copy of figure 3.23 in which the measurement at 11 cm form figure 3.24 is inserted)

The most likely cause of this difference is therefore considered to be the heat losses from the additional thermocouples. The additional thermocouples have been well insulated within the bed, but within the wall of the pot they were not ceramicly insulated as were the standard thermocouples. This could lead to extra heat losses to the wall for the additional thermocouples. It would also explain why the measured temperatures at the end of an experiment increase relatively to the gas inlet temperature when a series of experiments advances. This effect is clearly observed in figure 3.26, where the temperature of the additional thermocouples at 20 cm is much higher, relative to the gas inlet temperature, in the
last step change than in the first step change which is barely observed in the measurements by the standard thermocouples.

Figure 3.26, the effect of relative increase in measured temperature of the additional thermocouples during a series of experiments (copy of figure 3.23)

It can be explained by the fact that the longer the bed has been heated, the more heat will have been absorbed by the wall resulting in a higher wall temperature. The difference between the bed and the wall temperature will thus be smaller and the driving force for heat losses as well, resulting in a higher measured temperature.

This leads to another observation in the experimental data; the fact that the measured axial temperature profile is very strange. It is expected that with the bed losing heat to the wall, the temperature in the bottom of the bed will always be lower than the temperature at the top of the bed. The fact that this is not reflected in the measurements could not have been caused by heat losses through the thermocouple due to a lower wall temperature. If it were caused by these heat losses, the effect would not be present in the measurements of the standard thermocouples, as they are all insulated by a ceramic tube even within the wall. Furthermore, regarding the additional thermocouples, this type of heat loss would be expected to have a larger influence in the bottom of the bed where the wall has warmed up less, whereas from the experimental data it is concluded that the effect is larger at the top of the bed. The effect is also too high to be caused by the error in temperature measurement alone which is given as 0.4 and 0.1% of temperature for the additional and standard thermocouples respectively.

The reason for the difference in temperature is thought to arise from a combination of the error in measurement and the difference in local conditions around the thermocouple. This local difference can include the contact with the surrounding pellets and the difference in local flow conditions. Furthermore, it is thought to be influenced by the local radial gradient induced by the heat losses of the bed to the wall which is expected to differ locally. The combined effect of the differences mentioned above can not be evaluated but they can be dealt with when assumed constant during the experiments. When the effects are assumed to be constant they can be regarded as a constant systematic error which can be corrected for by subtracting the error from the measurement. In other words, shifting the experimental curves relative to each other on the temperature scale will not have any influence on the response of the thermocouple to the step change. The fact that a constant systematic error is found, is supported by the shape of the measured responses by the standard thermocouples. These can be shifted in such a way that for all the subsequent step changes an expected decreasing axial temperature profile is found as can be seen in figure 3.27.
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Figure 3.27 response curve of the standard thermocouple measurement after correction for a systematic error.

This kind of correction does not lead to good results for the measurements by the additional thermocouples, as the shape of the response changes during the subsequent experiments and with the depth in the bed. As was explained earlier, this is probably caused by changing heat losses through the thermocouple induced by the heating up of the wall.

It can be concluded that there will be a large error in the determination of the heat transfer coefficient from the irregularities and uncertainties in the experimental data alone. These can be divided into the effects of heat losses from the packed bed to the wall on the one hand and errors and uncertainties in the temperature measurement on the other. Considering the errors in the temperature measurement it can be concluded that the measurements of the standard thermocouples are much better than those by the additional thermocouples in which both synchronisation and heat losses play an important role.
3.5.2 Modelling results

Due to the aforementioned problems in the measurement, the below mentioned strategy has been used in modelling the packed bed, in order to arrive at estimates of the heat transfer coefficient. Considering the heat losses, first of all models have been used in which the influence of the heat effects does not have to be incorporated. When these models proved to be not accurate enough, it has been attempted to include the heat losses. This was done in such a manner that the one dimensional character of the model remained in order to avoid problems in validating the radial heat transfer.

With respect to the error in the temperature measurements, in obtaining estimates of the heat transfer coefficients, the data from the standard thermocouples have been used first. Although the information in the temperature response from the additional thermocouples inside a pellet are considered much more valuable and characteristic to the heat transfer, the problems of synchronisation and heat losses through the thermocouples are thus avoided. From this analysis it is concluded whether or not the model is viable. When this is the case, the model has been used to analyse the data from the additional thermocouples to get a better estimate of the heat transfer coefficient.

3.5.2.1 Quasi single-phase model

The quasi single-phase model is used to get a first estimate of the heat transfer coefficient without needing to refer to numerical modelling. In the quasi single-phase model the problem of the heat losses is neglected by looking at the top of the bed where the influence of the heat losses from the bed to the wall is expected to be absent. The heat losses in the burner section have been incorporated by taking a linear rise in gas inlet temperature as the boundary condition at the top of the bed. The response of the top of the bed according to the quasi single-phase model is for these boundary conditions given by equation 3.16 and 3.17.

As explained, the model was first used in combination with the measurements from the standard thermocouples. For the experimental temperature of the top of the bed, the standard thermocouple at the highest position in the pot 11 cm was used which lies effectively 5.5 cm under the top of the bed. In principle, the assumption in the quasi single-phase model that the gas temperature is equal to the gas inlet temperature is not valid at this position. However, it has the advantage that the effects of radiation by the burner and the influence of strong local varying conditions, which are expected to be found just underneath the top of the bed, are effectively dampened.

The effect of taking a standard thermocouple measurement instead of using the measurement by a thermocouple inside a pellet can be estimated by comparing the response times of both measurements, as is shown in figure 3.28 for a flow of 555 and 140 Nm³/hr respectively. It can be seen that the difference in response time is dependent on the process conditions. At best, the responses will be comparable and at worst the response of the standard thermocouple will be faster by some 10%. The analysis using the standard thermocouples will thus give an estimate of the overall heat transfer coefficient which will be too high but is considered valuable as a first estimate and as an indication of the viability of the model.

This has both advantages as well as disadvantages; as the thermocouple is not at the top of the bed the assumption of the gas temperature at that position being the same as at the top of the bed is not true. The standard thermocouple at 11 cm is used as experimental data because it showed the least systematic error and thus did not have to be adapted. A disadvantage is that as the thermocouple is not inside a pellet, it is not possible to attain a separate external and internal heat transfer coefficient from the experiments. However, the characteristic time of the response of the standard thermocouple and the thermocouple inside differed only a maximum of 10% as can be seen in figure 3.28 making the standard thermocouple measurements eligible for obtaining a quick estimate.
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To obtain the heat transfer coefficient, the model equation of 3.16 is fitted to the experimental curve by changing the value of the characteristic time \( t \) using a criteria. In this approach a linear fit of the gas inlet temperature has been used. An example of the result of a fit is shown figure 3.29

The fit in figure 3.29, and those of other experiments show a significant deviation after the initial rise in temperature. This is expected to be caused by the influence of the heat losses in the bed which will slow down the heating after the initial steep rise of the temperature. Considering the top of the bed these effects can be neglected, but as the measurements have been conducted significantly underneath the top, this effect will not be absent in the experimental data.

From the obtained value for the characteristic time, the overall heat transfer coefficient is calculated using equation 3.17, in which the interfacial heat transfer coefficient is replaced by the total heat transfer coefficient according to equation 3.31. In this approach no difference is made between the fines and the pellets, as the entire solid phase is characterised by a uniform average temperature. In the calculation of the heat transfer coefficient per unit area, the combined area of both pellets and fines is used assuming a spherical shape for the fines...
leading to the results for the heat transfer coefficient given in figure 3.30, 3.32 and appendix H.

![Graph](image)

**Figure 3.30**: dependency of overall heat transfer coefficient on bed voidage shown for a flow of 270 Nm$^3$/hr (lines are used to guide the eye)

The absolute value of the overall heat transfer coefficients obtained lies in the range from 20 - 120 W/m$^2$K. Although the overall heat transfer of a thermocouple and a pellet resemble, this does not mean that both the internal and external heat transfer coefficients are the same. It is therefore not useful to try and obtain a value for the (external) interfacial heat transfer coefficient by subtracting the influence of the internal heat transport. On the other hand the results can very well be used to see whether or not a trend can be discovered in the influence of the temperature, flow rate and bed voidage on the overall heat transfer coefficient.

In the experiments at a constant flow of 270 Nm$^3$/hr and varying bed voidage, a trend can be found that the heat transfer coefficient per unit area decreases with decreasing bed voidage. This influence of the bed voidage is the opposite of what was thought. However, these results are very likely to be heavily influenced by the assumptions made in the model. For one, the total interfacial area has been estimated on the basis of uniform spherical shaped fines. The influence of this assumption can be discarded, by looking at the volumetric heat transfer coefficient which is not based on the total interfacial area in the bed, but on the volume of the bed. It can be calculated from the interfacial heat transfer coefficient using equation 3.51 the result of which is given in figure 3.31.

![Graph](image)

**Figure 3.31**: the influence of the bed voidage on the volumetric heat transfer coefficient (lines are used to guide the eye)
The modification has made the trend in the influence of the bed voidage less clear, but there is still a distinct worsening of the heat transfer coefficient visible with decreasing bed voidage.

Another assumption influencing the heat transfer coefficient is the assumption that the top of the bed is measured, while in fact, the measurements have been conducted at a depth of 5.5cm. The influence of measuring at a certain depth in the bed is different depending on the voidage. At lower voidage, a measurement at the same depth will be more influenced as there will be more mass of the bed above it which will lead to a larger difference between the gas temperature at that depth in comparison with the gas inlet temperature.

With respect to the influence of temperature on the heat transfer coefficient, the results show no clear trend. The curve at a voidage of 0.37 shows practically no temperature dependency while the scatter in the curves of 0.41 and 0.39 is too large to draw any conclusion on the temperature dependency. A better indication for the influence of temperature on the heat transfer coefficient can be found in the results for different flow rates at a constant voidage of 0.41 given in figure 3.32.

![Figure 3.32, influence of temperature and flow on the overall heat transfer coefficient for a bed voidage of 0.41 (dotted lines denote the average over the temperature interval)](image)

From this figure, it cannot be concluded that there is trend for the heat transfer coefficient to slightly increase with increasing temperature as was found in literature and can be see figure 3.1. It can be observed that there is a trend for the heat transfer coefficient to increase with increasing flow. For the average heat transfer coefficient over the temperature interval this trend is shown in figure 3.33 as being approximately linear. The trend found by Kitaev (figure 3.1 c) is that the heat transfer coefficient is proportional with the flow rate to the power 0.7.
Using the quasi single-phase model, estimates for the heat transfer coefficient have been obtained. In this investigation it has been found that the overall heat transfer increases with increasing flow rate and increasing bed voidage, but no clear trend in the influence of temperature was observed. However, no decisive conclusion can be drawn from this analysis as the assumptions of the model are most likely to be invalid at the position of the measurement and will influence the estimate of the overall heat transfer coefficient. In this respect, it is referred to the paragraph on the Schuhmann model in which it is shown that the response of the solid phase at 5.5 cm in the bed is significantly different from the response at the top of the bed. It has been chosen therefore, not to use this analysis on the experimental data from the additional thermocouples, but to determine the effects of an improved model which would incorporate these effects.

3.5.2.2 Exact solution to the Schuhmann model

As was shown by the Schuhmann model, the response of the bed changes significantly with the depth in the bed. As measurements have been taken at multiple levels in the bed, the accuracy in the estimation of the heat transfer coefficient could be greatly improved if these different measurements were used in the analysis at the same time, using the solution to the model given in equations 3.18-3.19.

In the exact solution to the Schuhmann model, given by equations 3.23-3.27, heat losses, a rising gas inlet temperature and a non uniform initial bed temperature have not been included. However, it could be that the influence of the rise in gas inlet temperature is dampened by the bed and does not significantly alter the response of deeper layers in the bed. Furthermore, it could be assumed that the heat losses pose only an additional effect, lowering the temperatures in subsequent layers of the bed but not influencing the shape of the response. In this case the exact solution to the Schuhmann model would still be valid when the influence of the heat losses is subtracted from the measured response. This is done by regarding the dimensionless temperature response of the bed at each depth in the bed. That is to say, each response curve is made dimensionless using the initial and final temperature of that particular response. In the analysis using the classical Schuhmann model the standard thermocouple measurements have again been used in order to avoid problems of synchronisation and get a relatively quick estimate of the accuracy of the approach.
The results of such an analysis, of which an example is shown in figure 3.34, can be found in table 3.6. It was concluded that the heat losses and rise in gas inlet temperature can not be dealt with in this fashion. In the analysis, only the measurements of the first two thermocouples have been used, and even then it was not possible to get an acceptable fit leading to the conclusion mentioned above. When taking into account the large error in the fit procedure, no trends can be discovered in the results of the analysis by the exact solution to the Schuhmann model.

Table 3.6 results Schuhmann analysis

<table>
<thead>
<tr>
<th>flow (Nm³/hr)</th>
<th>T (K)</th>
<th>voidage</th>
<th>hᵣₑₚₑ(W/m²K)</th>
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</thead>
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<tr>
<td>270</td>
<td>723</td>
<td>0.41</td>
<td>1.7*10⁶</td>
</tr>
<tr>
<td>270</td>
<td>723</td>
<td>0.39</td>
<td>2.5*10⁶</td>
</tr>
<tr>
<td>270</td>
<td>1023</td>
<td>0.41</td>
<td>2.0*10⁶</td>
</tr>
<tr>
<td>555</td>
<td>723</td>
<td>0.41</td>
<td>2.3*10⁶</td>
</tr>
</tbody>
</table>

Figure 3.34, example of an analysis using the exact solution to the Schuhmann model for the first two standard thermocouples.
3.5.2.3 Numerical solution to the Schuhmann model

In order to incorporate the increase in gas inlet temperature during an experiment, it has been necessary to solve the Schuhmann model given by the set of equation 3.18 to 3.19 numerically in which the actual measured fluctuation in gas inlet temperature and flow were included. The numerical implementation is given in equations 3.61 and 3.66 for the fluid and solid phase respectively. It was further avoided having to incorporate heat losses into the model, by looking only at the thermocouple data from the top of the bed. In the top of the bed the influence of heat losses are considered absent according to the analysis given in paragraph 3.3.6.1. In order to get an estimate of the external heat transfer separate from the total heat transfer coefficient, the data from the thermocouple inside a pellet has been used of which the highest was located at 11 cm.

Before the numerical model was used, the data from the experiments has been tested by comparing the results of the model with the exact solution to the Schuhmann equation. For this comparison, the same boundary conditions have been used in the numerical model as were used in the exact solution to the Schuhmann model. From the result of this comparison, given in figure 3.35 it can be concluded that the numerical model is accurate.

![Figure 3.35, comparison between numerical and exact solution of the Schuhmann model](image)

It has to be noted here that in case of the exact solution to the Schuhmann model, all the thermophysical properties of both fluid and solid have been considered constant. In the numerical modelling of the experiments, these have been taken as dependent on the temperature. This results in an error induced by manner in which the specific heat is incorporated. However, this error has been investigated by using a heat balance over the bed. This induces an error in the order of 3%.

The model was used to analyse the experimental data from one of the additional thermocouples which measured the temperature inside a pellet and the gas inlet temperature from the measurements by the process control system. This meant that before the response data could be analysed, the problem of synchronisation and of the disagreement in absolute measured temperature had to be overcome. In order for the experimental data to be analysed using the numerical Schuhmann model, the initial solid and fluid temperatures had to be the same, and the solid temperature should be lower than the inlet gas temperature at all times during the experiment.

The experimental data showed a solid temperature which was much too low, sometimes even lower than the outlet gas temperature as was shown in figure 3.23. At first it was thought that
this was a constant systematic error and could be corrected for by shifting the entire curve so that the initial gas temperature was equal to the initial solid temperature. However, when this was done, the final gas temperature became higher than the inlet gas temperature which could never be predicted by the model and would disturb the fitting process. In order to be able to get any estimates of the heat transfer coefficient, the data had to be pre processed. This was done by making the temperature response of the thermocouple dimensionless in its own initial and end temperature and then make the response curve dimension bearing again by using the initial and end temperature of the gas curve. In this way, the characteristic shape of the curve and thus the estimate for the heat transfer coefficient would be left untouched while the beginning and end temperature were such that the model was able to come to a fit. It has to be noted that the effect which caused the non constant systematic error has not disappeared and is likely to influence the result significantly.

The synchronisation of the two curves is a difficulty which has been discussed in detail above. The effect of the synchronisation lay in the changing time lag of the response. By shifting the experimental curve of the gas inlet temperature relative to the measurement by the additional thermocouple, the time lag of the response could be increased or decreased. However, by looking at the sensitivities of the model, it was found that the time lag predicted by the model varied only in the order of a few seconds within the expected range of heat transfer coefficients (50 - 500 W/m\(^2\)K). The predicted time lag by the model was therefore used to synchronise the measurements by the process control system and by the personal computer.

The obtained fitted curves, of which 3.35 is an example, are mostly very good except that in some curves the beginning is not well fitted. This can be explained by the assumption of a uniform temperature in the bed at the beginning of the experiment while in reality the bed temperature was continuously rising. The initial temperature of the bed was therefore not static as was assumed, but dynamic. The fact that most curves showed a good fit, is in this case not a guarantee for the correctness of the model, as the experimental curve was scaled using the dimensionless response to fit the beginning and end temperature of the gas temperature. Furthermore, experimental effects on the time lag were discarded as well.

The results from the analysis of the experimental data for the dependency of flow and the temperature at a constant voidage of 0.41 are given in figure 3.37 and appendix H.
Results and discussion

In the measurements no trend can be discovered with respect to the temperature and therefore, the flow dependency has been investigated on the basis of the average heat overall transfer coefficient over the measured temperature range is given in figure 3.38 and appendix H.

In figure 3.38 it can be seen that there is a rising trend in overall heat transfer coefficient with higher flow rate, but at the highest flow rate, a much lower overall heat transfer coefficient is measured.

This could be explained by the fact that the overall heat transfer is influenced both by the internal and the external heat transfer according to equation 3.31. This relation can directly be
applied to the results of the overall heat transfer coefficient, as in the experiments on the flow dependency no fines were added to the bed. A reason for the unexpected lower value of the overall heat transfer could be that as the experiments at this flow rate, 700 Nm³/hr, were conducted only at lower temperatures (due to the restrictions on the pressure drop), the effective conductivity of the pellet might have been lower than in case of the experiments at 555 Nm³/hr.

In order to check this, an assumption had to be made on the value and temperature dependency of the effective conductivity, which was investigated in the previous chapter, where in the conducted experiments an effective conductivity of the pellet was found of around 0.3 W/mK. Using equation 3.30, an internal heat transfer coefficient is found of 165 W/m²K. As the internal and external heat transport is in series, the overall heat transfer coefficient can be found using equation 2.29. It is concluded from the results that this effective conductivity can not be applied to the results, as it implies that the maximum overall heat transfer coefficient obtainable is 165 W/m²K, while the results indicate that higher overall heat transfer coefficients have been obtained. Other values and trends have been found by Beer and Bratchikov. However, the effective conductivity found by Bratchikov is strongly temperature dependent, making it not apt to be used on an overall heat transfer coefficient which has been averaged over measurements at different temperatures. It has been decided to use the conductivity found by Beer which is approximately constant at a value of 0.6 W/mK. The results of this analysis are also shown in figure 3.38 using the right axis. The interfacial heat transfer coefficients found are in general high compared to literature and the interfacial heat transfer coefficient found at a flow of 555 Nm³/hr is clearly unrealistic. As both the effective conductivity as well as the overall heat transfer coefficient are subject to high errors, unrealistic results were to be expected.

The dependency of the heat transfer coefficient on the bed voidage has also been investigated and the results are given in figure 3.39 and appendix H. The heat transfer coefficient has again been given in terms of bed volume to avoid the problem of unknown interfacial area.

The result must be very much influenced by the error in the heat transfer determination as it seems unlikely that the heat transfer coefficient exhibits a minimum with respect to the

\[ \text{Heat transport in the packed bed} \]
voidage. Although it is unlikely, theories can be proposed to support this minimum value by assuming an effective usage of interfacial area which decreases with increasing flow combined with a better heat transfer at those area which are used for interfacial heat transport. It might very well be that the difference is within the standard deviation and thus no further explanations are sought.

In order to improve the accuracy of the estimate of the heat transfer coefficient, it has been investigated whether or not, the measurements of a thermocouple measurement deeper in the bed could be included in the analysis. To check this, the experimental data has been analysed twice; first on the basis of the measurement by thermocouple at 11 cm and then on the basis of the thermocouple at 20 cm. The results of this analysis are given in figure 3.40 and figure 3.41 respectively.

**Figure 3.40, analysis the experimental data on the basis of the response of the bed at 11 cm flow rate = 555 Nm$^3$/hr, bed voidage = 0.41, $h^{\text{tot}} = 264$ W/m$^2$K**

**Figure 3.41, analysis the experimental data on the basis of the response of the bed at 20 cm flow rate = 555 Nm$^3$/hr, bed voidage = 0.41, $h^{\text{tot}} = 121$ W/m$^2$K**

*Heat transport in the packed bed*
Results and discussion

It is concluded from this analysis that the assumption of negligible heat losses is certainly not valid for the position of the lower thermocouple (20 cm) as the model prediction on the basis of the measurement by the highest thermocouple predicts a much faster response of the lower thermocouple than was measured. When the model prediction is based on the measurement by the lower thermocouple, the predicted response of the highest thermocouple is much slower than was measured. Furthermore, the shape of model prediction for the position of the lower thermocouple does give a good resemblance of the measured response.

Summarising it can be said that the uncertainty in the beginning of the experiment considering the thermocouples inside a pellet combined with the heat losses through the thermocouple have made it impossible to get a reliable estimate of the overall heat transfer coefficient using the data from the top of the bed at which the response is expected to be least influenced by the heat losses to the wall. The estimate could have been improved by using the responses of thermocouples at different heights in the bed but this turned out not to be feasible because of the large influence of the heat losses to wall at deeper layers in the bed.

3.5.2.4 Including heat losses to the wall using the particle based model

In a final approach to increase the accuracy of the estimated heat transfer coefficient by using the temperature responses at deeper layers in the bed, it has been attempted to incorporate the heat losses to the wall in the model of the packed bed. In order to retain the one dimensional character of the model, it has been assumed that the heat losses occurred only through convective transport from the fluid phase which was assumed to be ideally mixed at each depth in the bed and conduction within the wall. The heat transport has been modelled by an effective conduction model. The heat transport within the wall has been modelled in detail as the heating of the wall is effectively a penetration process due to the high mass and low conductivity of the wall. The resulting equations for the wall and their numerically implementation are given in equations 3.48 to 3.50 and 3.75 to 3.79 respectively.

The effect of the internal heat transfer has been modelled as well by including the conduction within the pellet. This has been done to see if any of the observed phenomena in the response of the bed at deeper layers could have been caused by effects of the temperature profile in the pellets which had been underestimated by using an average solid temperature and an overall heat transfer coefficient. The equations of the internal particle heating are given in equations 3.34 to 3.36 and the numerical implementation in equations 3.67 to 3.71.

In this model, three extra unknown variables are introduced namely the convective heat transfer coefficient from the fluid to the wall, the heat transfer coefficient from the wall to the environment and the internal conductivity of the pellet. For the estimate of the internal conductivity, the estimate resulting from the experiments of the previous chapter were used. If necessary it could be replaced by estimates from literature. The estimate of the heat transfer coefficient from wall to the environment turned out not to be of very much importance as within the duration of the experiments the heat in the wall would effectively not penetrate, according to the model. In reality, the outside of the wall did heat up significantly. This could very well be caused by an underestimation of the conductivity of the wall in combination with the presence of damage (observed in the inside of the pot) or non homogeneity in the wall which has been confirmed by the finding of hot spots at the outer surface of the wall.

The heat transfer coefficient from the fluid to the wall has been determined by a fitting the experimental data. It was already mentioned that at the end of each experiment a quasi steady state is observed in which practically all the heat transferred from the gas is used in heating up the wall. In this situation, the heat transfer to and within the pellet can be considered as having no influence on the temperature profile in the bed. This makes it possible to obtain the effective heat transfer coefficient from the gas to the wall by requiring the correct prediction of the temperature profile in the quasi steady state, resulting in a value of 60 W/m²K from the fit given in figure 3.42.
Results and discussion

Figure 3.42, the fit of the effective heat transfer from gas to wall showing both experimental thermocouple measurement at 11, 20, 30, 40.5, 48 and 50 cm respectively (blue), as well as the model fits at the appropriate depth in the bed (red).

In this fit the experimental data has been corrected for systematic errors by assuming constant heat losses per unit of length of the bed as was done in figure 3.27 which limits the accuracy of this approach. The next step was to try and fit the rest of the curve by changing the value of the heat transfer coefficient. As can already be seen from 3.42, this required a relatively high heat transfer coefficient. In fact, in order to obtain a good fit for the top thermocouple, the interfacial heat transfer coefficient as well as the internal heat transfer had to be taken extremely large as can be seen in figure 3.43, effectively leading to a situation in which the gas is always in equilibrium with the solid phase.

Figure 3.43, attempted fit (red) of the experimental data (blue) using the particle based model including the heat losses at same positions as the previous figure.
It is concluded from this that the one dimensional modelling of wall effects over estimates the effects of the heat losses as measured in the centre of the pot. This can be explained by the fact that the penetration of the heat losses to the wall, estimated in figure 3.13, is not taken into account in the one dimensional model as any heat effects of the wall are immediately reflected in the heating of the pellets. It might have been possible to include the penetration effect by, as was done in the Schuhmann model, considering only a central section of the bed in which the heat losses are still considered only for the gas phase but related to the wall temperature by an effective conduction process as was used in the calculation of the penetration depth but no time has been available for the implementation.
Conclusions and recommendations

In the experiments, an overall heat transfer coefficient was found of order 200 W/m²K and an interfacial heat transfer coefficient of order 500 W/m²K, which lies in the range expected from literature. With respect to the dependency of the heat transfer coefficient on temperature, bed voidage and flow rate, no decisive conclusion could be drawn.

In order to arrive at these estimates, a hierarchy of models has been developed for the heating of a packed bed as found in the experimental set-up. On the one hand, simple models, for which an exact solution exist, were used first, to get a quick estimate of the heat transfer coefficient. On the other hand, highly detailed models, including the axial temperature profile in the bed, the temperature profile within both the particles and the wall, and the changing inlet flow conditions have been developed to determine the heat transfer coefficients more accurately.

From the experiments, it was found that heat losses to the wall have a decisive influence on the experimental conditions in the bed, which was not known in advance. Detailed analysis on the initial response of the bed has shown that approximately 55% of the heat transferred from the gas is used to heat the packed bed, the other 45% being used to increase the temperature of the mass of the wall. The overall heat losses in experiment amount to 70% as the relative heat losses increases during the experiment.

In the absence of measurements on the radial temperature profile, it has been necessary to develop models which reflected the heat losses in a one dimensional way, as no validation of a two dimensional model would have been possible. Two different approaches to the problem of the heat losses have been undertaken. In the first approach the heat losses were discarded by modelling a central section at the top of the bed, where it was estimated that the heat losses had not yet penetrated during the experiment. This approach has led to encouraging results for the top of the bed, however, for deeper parts of the bed, this assumption did not hold.

In order to describe the effects of heat losses in deeper layers of the bed, a model has been developed in which the heat transport to and within the wall was included. Limited by the one dimensional nature of the experimental data, the heat transferred to the wall has been modelled as coming only directly from the fluid phase. With this model it has been possible to accurately describe the quasi steady state occurring at the end of each experiment. However, the penetration of the heat losses proved to be too decisive to model the initial response of the bed in this manner.

Summarising, it can be said that, the large magnitude of the heat losses has severely hindered an accurate determination of the heat transfer coefficient in the pellet firing process.

For future work on the heat transfer and other heat effects in the pellet induration process, it can be concluded that the emphasis should be on the heat losses from the bed to the wall. The approach would be to both reduce the heat losses and at the same time measure them more accurately.

One of the ways to reduce the heat losses, will be the introduction of better insulation with both a lower conductivity as well as a lower specific heat. From experiences of one of the European partners in the pellet quality project, it is known that in this way the heat losses can reduced substantially. In a simple approach found in literature, the heat losses could be further reduced by first heating the packed bed until a constant temperature has been achieved, after which the bed is cooled by atmospheric air until the desired initial temperature has been reached. The thermal insulation will maintain a temperature roughly equal to the average bed temperature between the end and the beginning of the experiment, thereby reducing the heat losses to the wall. A disadvantage of this method is, that it would be a lot more time consuming.
Should a better insulation layer and pre heating of the wall, not suffice for a one dimensional model of the bed to be valid, a two dimensional model should be developed. In order to validate this model, it will be necessary to accurately measure the heat losses. The measurement of the heat losses should consist of both measurements in the wall as well as in the bed at different radial positions. It would suffice to do this only a limited number of layers in the bed, say two or three, to get an accurate estimate of the heat transfer coefficient.

For the temperature measurements it is recommended to use thermocouples inserted in a pellet as was done in the present work although the method should first be further tested. It has not yet been possible to accurately do this, due to presumed heat losses through the thermocouple wire and lack of synchrony in the logging with the process control system. According to the comparison of measurements with the standard thermocouples, these heat losses can be discarded adequately by using ceramic insulation all through the wall of the pot. The lack of synchrony can be effectively dealt with by logging the temperature of the top suction pyrometer using the personal computer.

Furthermore, it was found that the conductivity resulting from the experiments in the previous chapter, predicted a maximum overall heat transfer coefficient which was lower than the minimum overall heat transfer coefficient determined in the experiments on the packed bed. This could be caused by the inaccuracy in the analysis of the packed bed experiments, but it could also support the conclusion drawn in the previous chapter; that pellets used in the determination of the effective conductivity were not adequately sintered. For a decisive conclusion on this matter, it is recommended to determine the conductivity of the pellets used in the packed bed experiments experimentally.

Finally, on the part of the extensive, numerical modelling, it can be said that although it has not lead to a satisfactory description of the pilot plant at present, the frame work has been set up to analyse different dynamic process in the pot, given that the heat losses can be sufficiently reduced. These processes would not only be limited to the study of heat transfer but, by relative little adaptation of the model, mass transfer can be included as well. This would enable the analysis of the influence of the oxidation of magnetite and the combustion of coke breeze, present in the actual pellet firing process, as well as evaporation in the drying section of the induration process.
### 3.7 List of symbols

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<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tbody>
<tr>
<td>A</td>
<td>Surface area</td>
<td>[m²]</td>
</tr>
<tr>
<td>cₚ</td>
<td>Specific heat</td>
<td>[J/kg K]</td>
</tr>
<tr>
<td>d</td>
<td>Diameter</td>
<td>[m]</td>
</tr>
<tr>
<td>D</td>
<td>Diffusivity</td>
<td>[W/m² K]</td>
</tr>
<tr>
<td>h</td>
<td>Heat transfer coefficient</td>
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<tr>
<td>hᵣ</td>
<td>Interfacial heat transfer coefficient</td>
<td>[W/m² K]</td>
</tr>
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<td>hₜ</td>
<td>Overall heat transfer coefficient</td>
<td>[W/m² K]</td>
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<td>hᵥₜ</td>
<td>Overall volumetric heat transfer coefficient</td>
<td>[W/m³ K]</td>
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<td>Nᵤₜ</td>
<td>Nusselt number based on the particle diameter (hᵣ dp/A')</td>
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<tr>
<td>p</td>
<td>Pressure</td>
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<td>Pr</td>
<td>Prandtl number (cₚ μ/λ)</td>
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<td>R</td>
<td>Resistance to heat flow</td>
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<td>Temperature</td>
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<td>r</td>
<td>Radius/radial position</td>
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<td>Normal superficial velocity</td>
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<td>Density</td>
<td>[kg/m³]</td>
</tr>
<tr>
<td>T</td>
<td>Dimensionless time</td>
<td>[-]</td>
</tr>
<tr>
<td>τ</td>
<td>Characteristic time</td>
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<tr>
<td>Ω</td>
<td>Radius of the wall</td>
<td>[m]</td>
</tr>
<tr>
<td>ω</td>
<td>Radial position in the wall</td>
<td>[m]</td>
</tr>
</tbody>
</table>

**superscripts**

- sf: solid fluid interface
- fs: fluid phase in equation of solid phase
- f: fluid phase
- ff: fluid phase in equation of fluid phase
- s: solid phase
- ss: solid phase in equation of solid phase
- w: wall phase
- we: wall environment interface
- wf: wall fluid interface

**Heat transport in the packed bed**

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List of symbols

eff ................ effective
cond................ conduction
conv................ convection
external.......... external
internal.......... internal

subscripts
B ................ at grid point on the outer boundary of the solid
Bi ................ at grid point on the inner boundary of the wall
Bo ................ at grid point on the outer boundary of the wall
C ................ at grid point on the centre boundary of the solid
E ................ at grid point east of grid point P
e ................ at the interface between the control volumes of grid points P and E
I ................ at the last grid point before a boundary
i ................ at the interface between the control volumes of grid point I and the boundary
P ................ at the grid point under consideration
W ................ at the grid point west of grid point P
w ................ at the interface between the control volumes of grid points P and W
1 ................ boundary condition (t>0)
0 ................ initial condition (t=0)
3.8 References

28. 'Personal communication on difference between recorded PPI duration and actual duration on induration recipe no. 3', Hoogland, H., Hoogovens, (1999).
4. Overall conclusions and recommendations

4.1 Conclusions

In the analysis of heat transport in the pellet firing process, both the heat transport in a packed pellet bed and within the pellets individually have been investigated.

The experiments on the heat transport within the pellet can be described by a heat conduction process governed by an effective conductivity. In the experiments and effective conductivity was found of 0.3 W/mK at 700K which rises with increasing temperature to 0.7 W/mK at 1400K. These conductivities were determined with an accuracy of about 20%. Compared with the available literature, the conductivity found is relatively low and the difference cannot be accounted for by the error in the experiment. It is thought that the pellets in the experiments were not very well sintered which could explain the lower conductivity found.

In the analysis of the experiments on the effective conductivity, both the heat transport from the oven tube to the pellet surface and the transport within the pellet had to be included in the model of the experiment. Although the description of the heat transport to the pellet surface included both radiation and conduction, it was found that for temperatures below 700K, this heat transport was not modelled in sufficient detail to obtain an accurate estimate for the effective conductivity.

In the experiments on the heat transfer in a packed pellet bed, an overall heat transfer coefficient was found of order 200 W/m²K and an interfacial heat transfer coefficient of order 500 W/m²K, which lies in the range expected from literature. With respect to the dependency of the heat transfer coefficient on temperature, bed voidage and flow rate, no decisive conclusion could be drawn.

In order to arrive at these estimates, a hierarchy of models has been developed for the heating of a packed bed as found in the experimental set-up. On the one hand, simple models, for which an exact solution exist, were used first to get a quick estimate of the heat transfer coefficient. On the other hand, highly detailed models, including the axial temperature profile in the bed, the temperature profile within both the particles and the wall, and the changing inlet flow conditions have been developed to determine the heat transfer coefficients more accurately.

In the experiments it was found that approximately 45% of the heat from the gas was transferred to wall in the initial response of the bed and an average of 70% overall. In the absence of measurements on the radial temperature profile, it has been necessary to develop models which reflected the heat losses in a one dimensional way, as no validation of a two dimensional model would have been possible. Two different approaches to the problem of the heat losses have been undertaken. One in which the heat losses were discarded by looking at a central section at the top of the bed, where it was estimated that the heat losses had not yet penetrated during the experiment. In the other the heat transport to and within the wall was included. However, the heat losses have proven to be too decisive to arrive at an accurate estimation of the heat transfer coefficients.

4.2 Recommendations

In order to obtain more accurate estimates of the effective conductivity from the experiments in the tube oven, the axial temperature profile within the oven should be included in the analysis. Recommendations are given on how the model can be adapted to include this. It is further concluded that it would be better to consider a different experimental set-up, in which the heat transport to the pellet is far better than the heat transport within the pellet. Under such conditions, which are for example found when submerging a pellet in a well stirred bath of heated oil, more accurate estimates of the heat transfer coefficients can be obtained.
Based on the large influence of the heat losses as found in the experiments on the packed pellet bed, it has been decided to substantially reduce the heat losses. This is expected be achieved by using different insulation of lower density and conductivity, which has already been implemented in the experimental set up. The heat losses can further be reduced by a scheme in which the packed bed is subsequently pre heated and cooled, leaving the wall at a temperature between the initial and final temperature of the experiment.

Besides reducing the heat losses, it important that both the axial and the radial temperature profile will be measured during the experiment. The temperatures within the bed should be measured using thermocouples within a pellet as was done in the present work with some changes to the logging method and the insulation of the thermocouple within the wall. From the radial temperature profile measured, it can be assessed whether or not the insulation is adequate for allowing one dimensional modelling of the packed bed. Should this be the case, the models developed in the present work can used. Otherwise, a two dimensional model of the bed will be necessary in order to determine the heat transfer coefficients from the experiments. The radial temperature profile measured can then be used to validate this model.
A. Conduction from oven tube to pellet surface

The geometry of the pellet in a tube is modelled as two concentric spheres of uniform temperature. The steady state heat balance over a section enclosed by two concentric spheres results in the following differential equation:

$$0 = \frac{d}{dr} \left( r^2 \cdot \frac{dT}{dr} \right)$$

(A.1)

in which $r$ denotes the radial position from the centre of the inner sphere and $T$ denotes temperature. The general solution of this equation, together with the boundary condition of constant temperature of both spheres, will describe the temperature profile between the pellet and the oven wall.

$$T(r) = -\frac{C_1}{r} + C_2$$

(A.2)

$$T(r_{\text{pellet}}) = T_{\text{pellet}}$$

(A.3)

$$T(r_{\text{oven}}) = T_{\text{oven}}$$

(A.4)

in which $r_{\text{pellet}}$ denotes the radius of the pellet (inner sphere) and $r_{\text{oven}}$ the radius of the oven tube (outer sphere). The two constants $C_1$ and $C_2$ can be solved for,

$$C_1 = \frac{(T_{\text{pellet}} - T_{\text{oven}}) \cdot r_{\text{oven}} \cdot r_{\text{pellet}}}{r_{\text{pellet}} - r_{\text{oven}}}$$

(A.5)

$$C_2 = \frac{(T_{\text{pellet}} - T_{\text{oven}}) \cdot r_{\text{pellet}}}{r_{\text{pellet}} - r_{\text{oven}}} + T_{\text{oven}}$$

(A.6)

resulting in the description of the temperature profile:

$$T = -\frac{(T_{\text{pellet}} - T_{\text{oven}}) \cdot r_{\text{oven}} \cdot r_{\text{pellet}}}{r_{\text{pellet}} - r_{\text{oven}}} \cdot \frac{1}{r} + \frac{(T_{\text{pellet}} - T_{\text{oven}}) \cdot r_{\text{pellet}}}{r_{\text{pellet}} - r_{\text{oven}}} + T_{\text{oven}}$$

(A.7)

The steady state heat flow at the surface of the pellet can now be calculated by using Fick's law:

$$\phi_q = A_{\text{pellet}} \cdot \lambda_{\text{air}} \cdot \frac{dT}{dr}$$

(A.8)

$$\frac{dT}{dr} = \frac{(T_{\text{pellet}} - T_{\text{oven}}) \cdot r_{\text{oven}} \cdot r_{\text{pellet}}}{r_{\text{pellet}} - r_{\text{oven}}} \cdot \frac{1}{r^2}$$

(A.9)

$$\phi_q = A_{\text{pellet}} \cdot \lambda_{\text{air}} \cdot \frac{r_{\text{oven}} \cdot r_{\text{pellet}}}{r_{\text{pellet}} - r_{\text{oven}}} \cdot \frac{1}{r^2} \cdot (T_{\text{oven}} - T_{\text{pellet}})$$

(A.10)

wherein $\phi_q$ denotes the heat flow, $A_{\text{pellet}}$ the surface of the pellet and $\lambda_{\text{air}}$ the conductivity of the air in between the surface of the pellet and the oven tube. This final equation has the form of Newton's law of cooling so the transfer coefficient of the conduction process, related to the pellet surface area, can be expressed as:

$$h_{\text{conduction}} = \frac{\lambda_{\text{air}} \cdot r_{\text{oven}}}{(r_{\text{pellet}} - r_{\text{oven}}) \cdot r_{\text{pellet}}}$$

(A.11)
Appendix B

B. Analysis of resistance to heat transport for a pellet in a tube oven

references refer to chapter 2

process conditions

\[ T := 200 \text{K}, 250 \text{K}, \ldots, 1400 \text{K} \]

constants

\[ \sigma := 5.67 \times 10^{-8} \frac{\text{W}}{\text{m}^2\cdot\text{K}^4} \]

Stefan-Boltzman constant \hspace{1cm} (B.2)

temperature independant properties

\[ r_o := 11.5 \times 10^{-3} \text{m} \]

oven radius \hspace{1cm} (B.3)

\[ r_p := 6.1 \times 10^{-3} \text{m} \]

pellet radius \hspace{1cm} (B.4)

\[ \lambda_p := 0.1 \frac{\text{W}}{\text{m} \cdot \text{K}} \]

pellet conductivity \hspace{1cm} (B.5)

temperature dependent properties

\[ \varepsilon_p(T) := 5.625 \times 10^{-5} \frac{T}{K} + 7.931 \times 10^{-1} \]

pellet emissivity \hspace{1cm} (B.6)

\[ \varepsilon_o(T) := 1.1365 \times 5 \times 10^{-4} \frac{T}{K} \]

oven wall emissivity \hspace{1cm} (B.7)

\[ \eta_{\text{air}}(T) := \left( 3.30512 \times 10^{-8} \frac{T}{K} + 1.0534 \times 10^{-5} \right) \text{Pa}\cdot\text{s} \]

viscosity of air \hspace{1cm} (B.8)

\[ c_{\text{p,air}}(T) := \left( 0.208684 \frac{T}{K} + 9.29437 \times 10^{-1} \right) \frac{\text{J}}{\text{kg}\cdot\text{K}} \]

heat capacity of air \hspace{1cm} (B.9)

\[ \rho_{\text{air}}(T) := 3.4179 \times 10^{-3} \frac{\text{T}^2}{\text{K}} \left( 0.995051 \right) \frac{\text{kg}}{\text{m}^3} \]

density of air \hspace{1cm} (B.10)

\[ a_{\text{air}}(T) := \left( 2.317641 \times 10^{-3} \frac{T}{K} - 6.26554 \times 10^{-5} \right) \frac{\text{m}^2}{\text{K}} \]

heat diffusivity of air \hspace{1cm} (B.11)

\[ \lambda_{\text{air}}(T) := \left( a_{\text{air}}(T) \cdot \rho_{\text{air}}(T) \cdot c_{\text{p,air}}(T) \right) \]

heat conductivity of air \hspace{1cm} (B.12)

heat transfer coefficients (related to pellet surface)

Conduction, radiation and internal

\[ h_{\text{external}}(T) := \frac{\lambda_{\text{air}}(T) \cdot r_o}{(r_o - r_p) \cdot r_p} \]

heat transfer coefficient for conduction through air from oven wall to pellet surface (see appendix A) \hspace{1cm} (B.13)

\[ h_{\text{radiation}}(T) := \frac{4 \sigma \cdot (T)^3}{1 + \left( \frac{1 - \varepsilon_p(T)}{\varepsilon_p(T)} + \frac{(r_p)^2}{(r_o)^2} \cdot \frac{1 - \varepsilon_o(T)}{\varepsilon_o(T)} \right)} \]

heat transfer coefficient for radiation from oven wall to pellet \hspace{1cm} (B.14)

Analysis of resistance to heat transport for a pellet in a tube oven 100
Appendix B

Appendix B

\[ h_{\text{internal}}(p) = \frac{\pi^2 \lambda_p}{3 \cdot r_p} \]

heat transfer coefficient for conduction within the pellet \(^{15}\) (B.15)

Natural convection

![Diagram of natural convection with a model for calculation of free convection: pellet and wall are modelled as vertical flat plates of height \(2r_{\text{pellet}}\); the wall being 50 K higher in temperature than the pellet.]

Figure (B.1), model for calculation of free convection; pellet and wall are modelled as vertical flat plates of height \(2r_{\text{pellet}}\); the wall being 50 K higher in temperature than the pellet

\[ T_{\text{wall}}(T) := (T - 25 \text{ K}) \]

wall temperature \(^{(B.16)}\)

\[ T_{\text{pellet}}(T) := (T + 25 \text{ K}) \]

pellet temperature \(^{(B.17)}\)

\[ \Delta \rho(T) := \rho_{\text{air}}(T_{\text{wall}}(T)) - \rho_{\text{air}}(T_{\text{pellet}}(T)) \]

difference in density near oven wall and pellet surface \(^{(B.18)}\)

\[ v_{\text{air}}(T) := \frac{\eta_{\text{air}}(T)}{\rho_{\text{air}}(T)} \]

kinetic viscosity of air \(^{(B.19)}\)

\[ x := r_p \]

distance between the pellet and the oven wall \(^{(B.20)}\)

\[ \text{Gr}(T) := \frac{\left(3 \cdot \rho \cdot \Delta \rho(T)\right)}{v_{\text{air}}(T)^2 \cdot \rho_{\text{air}}(T)} \]

Graetz number \(^{(B.21)}\)

\[ \text{Pr}(T) := \frac{v_{\text{air}}(T)}{a_{\text{air}}(T)} \]

Prandtl number \(^{(B.22)}\)

\[ \text{Nu}(T) := 0.52 \left(\frac{\text{Gr}(T) \cdot \text{Pr}(T)}{1} \right)^{\frac{1}{4}} \]

Relationship for Nusselt number of free convection \(^{9}\) \(^{(B.23)}\)

\[ h_{\text{free-conv}}(T) := \frac{\text{Nu}(T) \cdot \lambda_{\text{air}}(T)}{2 \cdot r_p} \]

heat transport coefficient for free convection \(^{(B.24)}\)
N.B. Gr*Pr < $10^4$ so officially the equation does not hold; the driving force of the process is too small. The result will probably be an overestimation.

Resistances to heat flow based on the heat transfer coefficients

\[
\text{conduction}(T) := \frac{1}{h_{\text{external}}(T)} \\
\text{radiation}(T) := \frac{1}{h_{\text{radiation}}(T)} \\
\text{internal}(T, λ_p) := \frac{1}{h_{\text{internal}}(λ_p)} \\
\text{free_conv}(T) := \frac{1}{h_{\text{free_conv}}(T)} \\
\text{external}(T) := \frac{1}{\frac{1}{\text{conduction}(T)} + \frac{1}{\text{radiation}(T)}}
\]

Resistance of conduction through the air \(\text{eq. (B.25)}\)
Resistance of radiation from the oven wall \(\text{eq. (B.26)}\)
Resistance of conduction with in the pellet \(\text{eq. (B.27)}\)
Resistance of free convection \(\text{eq. (B.28)}\)
Total resistance to heat flow from the oven to the centre of the pellet \(\text{eq. (B.29)}\)

(free convection has been neglected)
C. Analysis of the temperature profile between oven tube and pellet

*references refer to chapter 2*

**Process conditions**

\[ T := 500 \text{K}, 550 \text{K}.. 1400 \text{K} \]

**Properties of air**

*source: data companion (1991)*

\[ c_{p\text{-air}}(T) := \left(0.208684 \frac{T}{K} + 9.29437 \times 10^2\right) \frac{J}{\text{kgK}} \]

\[ \rho_{\text{air}}(T) := 3.41790 \times 10^2 \left(\frac{T}{K}\right)^{-0.995051} \frac{\text{kg}}{\text{m}^3} \]

\[ a_{\text{air}}(T) := \left(2.31764 \times 10^7 \frac{T}{K} - 6.26554 \times 10^5\right) \frac{\text{m}^2}{\text{s}} \]

\[ \lambda_{\text{air}}(T) := \left(a_{\text{air}}(T) \rho_{\text{air}}(T) c_{p\text{-air}}(T)\right) \]

**Properties of pellet**

\[ \rho_{\text{pellet}} := 3280 \frac{\text{kg}}{\text{m}^3} \]

\[ c_{p\text{-pellet}} := 900 \frac{J}{\text{kgK}} \]

\[ \lambda_{\text{pellet}} := 0.1 \frac{\text{W}}{\text{mK}}, 0.2 \frac{\text{W}}{\text{mK}}, 1 \frac{\text{W}}{\text{mK}} \]

\[ a_{\text{pellet}}(\lambda_{\text{pellet}}) := \frac{\lambda_{\text{pellet}}}{\rho_{\text{pellet}} c_{p\text{-pellet}}} \]

**Penetration time in air**

Transport of heat from oven wall to pellet surface.

An estimation of the penetration time both in the air and the pellet is \( F_0 = 0.1 \)

\[ F_0 := 0.1 \]

\[ d_{\text{gap}} := 0.006 \text{m} \]

\[ t_{\text{air}}(T) := F_0 \frac{(d_{\text{gap}})^2}{a_{\text{air}}(T)} \]

**Penetration time in the pellet**

Transport of heat from pellet surface to the centre.

\[ d_{\text{p}} := 0.012 \text{m} \]

\[ t_{\text{pellet}}(T, \lambda_{\text{pellet}}) := F_0 \frac{(d_{\text{p}})^2}{a_{\text{pellet}}(\lambda_{\text{pellet}})} \]
D. Analysis of the heat losses through the thermocouple wire

A thermocouple consists of two wires embedded in insulation around which a steel sheath is placed.

Thermocouple geometry

\[ r_{tk} := 1 \cdot 10^{-3} \text{ m} \]
\[ L_{tk} := 0.01 \text{ m}, 0.02 \text{ m}, 0.5 \text{ m} \]
\[ A_{\text{sheath}} := \pi \cdot (r_{tk})^2 \left[ 1 - (0.88)^2 \right] \]
\[ A_{\text{sheath}} = 7.08 \times 10^{-7} \cdot \text{m}^2 \]
\[ A_{\text{wire}} := 2\pi \cdot (0.2 \cdot r_{tk})^2 \]
\[ A_{\text{wire}} = 2.51 \times 10^{-7} \cdot \text{m}^2 \]
\[ A_{\text{insulation}} := \left[ \pi \cdot (r_{tk})^2 \right] (0.88)^2 - A_{\text{wire}} \]
\[ A_{\text{insulation}} = 2.18 \times 10^{-6} \cdot \text{m}^2 \]
\[ r_{\text{pellet}} := 0.006 \text{ m} \]
\[ A_{\text{pellet}} := 4\pi \cdot (r_{\text{pellet}})^2 \]

Conductivity

Estimated from conductivity of steel and alumina

\[ \lambda_{\text{wire}} := 80 \frac{\text{W}}{\text{m-K}} \]
\[ \lambda_{\text{sheath}} := 80 \frac{\text{W}}{\text{m-K}} \]
\[ \lambda_{\text{insulation}} := 5 \frac{\text{W}}{\text{m-K}} \]

Heat transfer coefficients

A fully developed temperature profile is assumed in the thermocouple. The separate parts of the thermocouple (wire, isolation and sheath) are modelled as independent infinite plates.

\[ h_{\text{sheath}} (L_{tk}) := \frac{\lambda_{\text{sheath}}}{L_{tk}} \]
\[ h_{\text{wire}} (L_{tk}) := \frac{\lambda_{\text{wire}}}{L_{tk}} \]
\[ h_{\text{insulation}} (L_{tk}) := \frac{\lambda_{\text{insulation}}}{L_{tk}} \]
Appendix D

\[ UA(L_{tk}) := h_{wire}(L_{tk}) \cdot A_{wire} + h_{sheath}(L_{tk}) \cdot A_{sheath} + h_{insulation}(L_{tk}) \cdot A_{insulation} \]  
\[ UA(0.5 \text{ m}) = 1.75 \times 10^{-4} \text{ m}^{-1} \cdot \text{K}^{-1} \cdot \text{W} \]  
\[ \text{total heat transfer coefficient} \]  

Heat flows through thermocouple

\[ T_{ambient} := 298 \text{ K} \]  
\[ \Delta T_{tk}(T) := T - T_{ambient} \]  
\[ Q_{tk}(T) := UA(0.5 \text{ m}) \cdot \Delta T_{tk}(T) \]  
\[ \text{total heat flow from the pellet through the thermocouple assuming rest of the wire is completely isolated} \]  

Heat flows through pellet

\[ \varepsilon_p(T) := 5.625 \times 10^{-5} \frac{T}{K} + 7.931 \times 10^{-1} \]  
\[ \varepsilon_o(T) := 1.1365 - 5.10^{-4} \frac{T}{K} \]  
\[ \sigma := 5.67 \times 10^8 \frac{\text{W}}{\text{m}^2 \cdot \text{K}^4} \]  
\[ r_{oven} := 23 \times 10^{-3} \text{ m} \]  

\[ \Delta T_{pellet} := 50 \text{ K} \]  
\[ Q_{pellet}(T) := \frac{4 \sigma (T)^2 \cdot \Delta T_{pellet} \cdot A_{pellet}}{1 + \left( \frac{1 - \varepsilon_p(T)}{\varepsilon_p(T)} \right) + \left( \frac{r_{pellet}}{r_{oven}} \right)^2 \left( 1 - \varepsilon_o(T) \right) \varepsilon_o(T)^2} \]  
\[ \text{heat flow to pellet surface through radiation} \]  

Heat loss compared to heat input

\[ \text{heat loss (T)} := \frac{Q_{tk}(T)}{Q_{pellet}(T)} \cdot 100 \]  
\[ T := 500 \text{ K}, 550 \text{ K}, 1400 \text{ K} \]  
\[ \text{temperature range} \]  

This is presumably an exaggeration as a portion of the heat that leaves the thermocouple is supplied by the ceramic tubes. Furthermore, there is a radiative heat flow to the bare part of the thermocouple

Radiative flux to bare part of thermocouple

\[ L_{bare} := 0.006 \text{ m} \]  
\[ \text{part of thermocouple which is assumed bare} \]
Appendix D

\( \varepsilon_{tk} := 0.15 \)

\( h_{\text{radiation bare}}(T) := \frac{4 \sigma \cdot (T)^3}{1 + \varepsilon_{tk} \cdot r_{tk} + \varepsilon_{tk} \cdot r_{oven} \cdot \varepsilon_{o(T)}} \)

thermocouple emissivity (taken as polished steel) \( (D.28) \)

heat transfer coefficient for radiation transport to bare section \( (D.29) \)

\( \Delta T_{\text{bare}} := 50 \text{ K} \)

temperature difference between thermocouple and oven \( (D.30) \)

\( A_{\text{bare}} := 2 \pi \cdot r_{tk} \cdot L_{\text{bare}} \)

Area on bare section available for radiation transport

\( Q_{\text{radiation bare}}(T) := h_{\text{radiation bare}}(T) \cdot A_{\text{bare}} \cdot \Delta T_{\text{bare}} \)

Total radiative flux to thermocouple \( (D.31) \)

\( Q_{\text{theoretical}}(T) := \frac{Q_{tk}(T) \cdot Q_{\text{radiation bare}}(T)}{Q_{\text{pellet}}(T)} \cdot 100 \)

Percentage of heat lost in experiment according to theory \( (D.32) \)

Conductive flow on the basis of the thermocouple measurement in the insulation

Experimentally measured temperature difference between pellet and insulation

\( \Delta T(T) := \left[ -0.0002 \left( \frac{T}{273} \right)^2 + 0.1137 \left( \frac{273 - T}{K} \right) + 67.07 \right] \text{ K} \) \( (D.33) \)

\( UA(0.02m) = 4.386 \times 10^{-3} \text{ K}^{-1} \text{W} \) \( (D.34) \)

\( Q_{\text{conduction}}(T) := \Delta T(T) \cdot UA(0.02) \) \( (D.35) \)

\( \text{thermocouple}(T) := \frac{Q_{\text{conduction}}(T) \cdot Q_{\text{radiation bare}}(T)}{Q_{\text{pellet}}(T)} \cdot 100 \)

Percentage of heat lost in experiment. \( (D.36) \)
E. Results of the experiments on internal pellet heat transport

Table E.1, summary of results for model with radiation and conduction

<table>
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<tr>
<th>experiment no</th>
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<th>27</th>
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<th>31</th>
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<tbody>
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<td>( \lambda ) (W/mK)</td>
<td>0.58</td>
<td>0.79</td>
<td>0.31</td>
<td>0.34</td>
<td>0.31</td>
<td>0.28</td>
<td>0.27</td>
<td>0.24</td>
<td>0.30</td>
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<tr>
<td>( T_0 ) (K)</td>
<td>529.8</td>
<td>579.8</td>
<td>629.9</td>
<td>695.7</td>
<td>742.3</td>
<td>798.8</td>
<td>830.4</td>
<td>866.3</td>
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<tr>
<td>( T_{\text{final}} ) (K)</td>
<td>579.6</td>
<td>629.6</td>
<td>695.6</td>
<td>742.1</td>
<td>797.3</td>
<td>830.0</td>
<td>866.2</td>
<td>910.8</td>
<td>954.3</td>
</tr>
<tr>
<td>( T_{\text{average}} ) (K)</td>
<td>554.7</td>
<td>604.7</td>
<td>662.8</td>
<td>718.9</td>
<td>769.8</td>
<td>814.4</td>
<td>858.3</td>
<td>898.5</td>
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</table>

<table>
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<tr>
<td>( \lambda ) (W/mK)</td>
<td>0.27</td>
<td>0.27</td>
<td>0.28</td>
<td>0.33</td>
<td>0.39</td>
<td>0.43</td>
<td>0.39</td>
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<tr>
<td>( T_0 ) (K)</td>
<td>954.9</td>
<td>1005.2</td>
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<td>1116.5</td>
<td>1172.1</td>
<td>1213.9</td>
<td>1248.1</td>
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<td>1062.7</td>
<td>1116.0</td>
<td>1172.0</td>
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<tr>
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<td>1033.9</td>
<td>1089.4</td>
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<td>1192.8</td>
<td>1231.0</td>
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Table E.2, summary of results for model with radiation only

<table>
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<tbody>
<tr>
<td>( \lambda ) (W/mK)</td>
<td>n/a</td>
<td>n/a</td>
<td>0.49</td>
<td>0.50</td>
<td>0.40</td>
<td>0.28</td>
<td>0.28</td>
<td>0.20</td>
<td>0.35</td>
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<tr>
<td>( T_0 ) (K)</td>
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<td>579.8</td>
<td>629.9</td>
<td>695.7</td>
<td>742.3</td>
<td>798.8</td>
<td>830.4</td>
<td>866.3</td>
<td>911.0</td>
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<td>( T_{\text{final}} ) (K)</td>
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<td>695.6</td>
<td>742.1</td>
<td>797.3</td>
<td>830.0</td>
<td>866.2</td>
<td>910.8</td>
<td>954.3</td>
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<td>( T_{\text{average}} ) (K)</td>
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<td>662.8</td>
<td>718.9</td>
<td>769.8</td>
<td>814.4</td>
<td>858.3</td>
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<th>40</th>
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<td>0.28</td>
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<td>0.38</td>
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<td>1172.1</td>
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<tr>
<td>( T_{\text{final}} ) (K)</td>
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<td>1062.7</td>
<td>1116.0</td>
<td>1172.0</td>
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<td>1248.0</td>
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<tr>
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<td>1144.2</td>
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Table E.3, summary of results for model with constant surface temperature

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<th>( T ) (°C)</th>
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<th>( C_p ) (J/kgK)</th>
<th>( \lambda ) (W/mK)</th>
<th>( T ) (°C)</th>
<th>1/( \tau )</th>
<th>( C_p ) (J/kgK)</th>
<th>( \lambda ) (W/mK)</th>
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<tr>
<td>632.7</td>
<td>0.01031</td>
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</tbody>
</table>

Analysis of the radial penetration of heat losses in the bed
Appendix F

F. Analysis of the radial penetration of heat losses in the bed

references refer to chapter 3

Process conditions

\[ T = 500 \text{K}, 525 \text{K}, 1300 \text{K} \]

\[ \phi v_n = 150 \frac{m^3}{hr}, 175 \frac{m^3}{hr}, 1000 \frac{m^3}{hr} \]

\[ \varepsilon = 0.37, 0.38, 0.41 \]

\[ \text{time} = 0 \text{s}, 10 \text{s}, 3600 \text{s} \]

Properties of fluid

\[ c_{p \text{air}}(T) = \left( 0.20868 \frac{T}{K} + 9.29437 \times 10^{-2} \right)^T \]

\[ \rho_{\text{air}}(T) = 3.417901 \left( \frac{T}{K} \right)^{0.995051} \]

\[ a_{\text{air}}(T) = 2.3176410^{-7} \left( \frac{T}{K} \right)^{6.2655410^{-5}} \]

\[ \lambda_{\text{air}}(T) = \left( a_{\text{air}}(T) \cdot \rho_{\text{air}}(T) \cdot c_{p \text{air}}(T) \right) \]

Properties of the bed

\[ \rho_{\text{solid}} = 3280 \frac{kg}{m^3} \]

\[ \rho_{\text{bed}}(T, \varepsilon) = (1 - \varepsilon) \cdot \rho_{\text{solid}} + \varepsilon \cdot \rho_{\text{air}}(T) \]

\[ c_{p \text{bed}} = 1000 \frac{J}{kg \cdot K} \]

\[ R_{\text{bed}} = 0.17 \text{m} \]

Estimation of effective radial thermal conductivity of the packed bed

\[ v_{\text{inter}}(\phi v_n, \varepsilon, T) = \phi v_n \cdot \frac{\rho_{\text{air}}(273 \text{K})}{\rho_{\text{air}}(T)} \cdot \frac{1}{\varepsilon \cdot \pi \cdot R_{\text{bed}}^2} \]

\[ \lambda_{\text{r}}(\phi v_n, \varepsilon, T) = \left[ 25 \frac{\text{S}}{\text{m}} \cdot v_{\text{inter}}(\phi v_n, \varepsilon, T) + 25 \right] \lambda_{\text{air}}(T) \]

\[ a_{\text{r}}(\phi v_n, \varepsilon, T) = \frac{\lambda_{\text{r}}(\phi v_n, \varepsilon, T)}{\rho_{\text{bed}}(T, \varepsilon) \cdot c_{p \text{bed}}} \]
Estimation of radial heat penetration of the packed bed

\[
d_{\text{pen}}(\phi_{v,n,e}, T, \text{time}) := \sqrt{a_{r}(\phi_{v,n,e}, T)} \cdot \pi \cdot \text{time}
\]

estimation of penetration depth \(^{20}\) \(\text{F.16}\)

conditions \((\phi_{v,n,e}, T, \text{time}) := \frac{d_{\text{pen}}(\phi_{v,n,e}, T, \text{time})}{R_{\text{bed}}} \cdot 100\) penetration relative to the diameter of the bed \(\text{F.17}\)
Appendix G

G. Analysis of the influence of heat losses on the flow

*references refer to chapter 3*

Variable conditions in the bed

\[
\begin{align*}
\phi_{\text{mtot}} &:= 0.01, 0.02, 0.02^2 \text{ kg s}^{-1} \text{ s} \\
\tau &:= 0, 0.01, 0.16 \text{ m} \\
\varepsilon &:= 0.36, 0.37, 0.41 \\
T &:= 300 \text{ K}, 325 \text{ K}, 1400 \text{ K} \\
\Delta T &:= 0, 1, 100 \text{ K} \\
\end{align*}
\]

Different dimension in the bed

\[
\begin{align*}
d_{\text{pel}} &:= 0.012 \text{ m} \\
R_{\text{pot}} &:= 0.17 \text{ m} \\
A_1(r) &:= \pi r^2 \\
A_2(r) &:= \pi \left( R_{\text{pot}}^2 - r^2 \right) \\
\end{align*}
\]

Calculation of pressure drop in each section using Ergun equation

\[
\begin{align*}
\alpha(\varepsilon) &:= 150 \left( 1 - \varepsilon \right)^2 \frac{1}{\varepsilon^3} \frac{d_{\text{pel}}^2}{d} \\
\beta(\varepsilon) &:= 1.75 \left( 1 - \varepsilon \right)^{-1} \frac{1}{\varepsilon^2} \frac{d_{\text{pel}}}{d} \\
\rho_2(T) &:= 3.4179010^{2.7} \frac{T}{K} \left( -0.995051 \right) \frac{\text{kg}}{\text{m}^3} \\
\eta_2(T) &:= \left( 3.3051210^8 \frac{T}{K} + 1.0534010^{-5} \right) \text{ Pa s} \\
\rho_1(T, \Delta T) &:= 3.4179010^{2.7} \frac{T}{K} \left( -0.995051 \right) \frac{\text{kg}}{\text{m}^3} \\
\eta_1(T, \Delta T) &:= \left[ 3.3051210^8 \frac{T}{K} + 1.0534010^{-5} \Delta T \right] \text{ Pa s} \\
\end{align*}
\]

Calculation of resultant flows in each section using Ergun equation

Assuming equal pressure drops over each of the sections, the Ergun equation of the inner and the outer section (A1 and A2) and be set equal to each other. This results in a quadratic equation:

\[
A_1 \phi_{\text{m1}} + A_2 \phi_{\text{m1}} + C = 0
\]

in which \( \phi_{\text{m1}} \) denotes the mass flow rate through the inner section of the bed.
Appendix G

Different terms in equation G.16

\[ A(r, ε, T, ΔT) := \beta(ε) \left( \frac{-1}{ρ(T) A_2(r)^2} + \frac{1}{ρ(T, ΔT) A_1(r)^2} \right) \]

constant in quadratic term (G.17)

\[ B(φ_{mtot}, r, ε, T, ΔT) := \frac{α(ε) η_1(T, ΔT)}{ρ(T, ΔT) A_1(r)} + \frac{α(ε) η_2(T)}{ρ(T) A_2(r)} + \frac{2φ_{mtot} β(ε)}{ρ(T) A_2(r)^2} \]

constant in linear term (G.18)

\[ C(φ_{mtot}, r, ε, T, ΔT) := \frac{-α(ε) η_2(T)}{ρ(T) A_2(r)} - \frac{φ_{mtot} β(ε)}{ρ(T) A_2(r)^2} \]

constant term (G.19)

Determinant

\[ D(φ_{mtot}, r, ε, T, ΔT) := \left( B(φ_{mtot}, r, ε, T, ΔT) \right)^2 - 4A(r, ε, T, ΔT) C(φ_{mtot}, r, ε, T, ΔT) \]

(G.20)

Solution of equation G.16

\[ φ_{ml}(φ_{mtot}, r, ε, T, ΔT) := \frac{-B(φ_{mtot}, r, ε, T, ΔT) + \sqrt{D(φ_{mtot}, r, ε, T, ΔT)}}{2A(r, ε, T, ΔT)} \]

(G.21)

Mass flux through section A1 relative to the average mass flux through the bed

\[ \xi(φ_{mtot}, r, ε, T, ΔT) := \frac{φ_{ml}(φ_{mtot}, r, ε, T, ΔT)}{A_1(r)} \]

(G.22)
H. Results of packed bed heat transport experiments

Table G.1, results numerical Schuhmann and Quasi single-phase models

<table>
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</tr>
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<td>T_c (K)</td>
</tr>
<tr>
<td>-----</td>
</tr>
<tr>
<td>exp11</td>
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<td>exp19</td>
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<td>exp20</td>
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<tr>
<td>avg.</td>
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</tbody>
</table>

Results of packed bed heat transport experiments 112
Table G.2, results numerical Schuhmann and Quasi single-phase models
(mass of pellets: 84.3 kg, mass of fines: 4.5 kg, bed voidage: 0.39, free space above the bed: 5 cm)

<table>
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<tr>
<th>exp</th>
<th>$T_0$ (K)</th>
<th>$T_{final}$ (K)</th>
<th>$T_{avg}$ (K)</th>
<th>flow (Nm³/hr)</th>
<th>$h_{int}$ (W/m²K)</th>
<th>$h_{int}^t$ (W/m²K)</th>
<th>$h_{int}^v$ (W/m²K)</th>
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<tbody>
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<td>avg.</td>
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<td>793</td>
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Table G.3, results numerical Schuhmann and Quasi single-phase models
(mass of pellets: 84.3 kg, mass of fines: 4.5 kg, bed voidage: 0.39, free space above the bed: 4.5 cm)

<table>
<thead>
<tr>
<th>exp</th>
<th>$T_0$ (K)</th>
<th>$T_{final}$ (K)</th>
<th>$T_{avg}$ (K)</th>
<th>flow (Nm³/hr)</th>
<th>$h_{int}$ (W/m²K)</th>
<th>$h_{int}^t$ (W/m²K)</th>
<th>$h_{int}^v$ (W/m²K)</th>
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Table G.3, results numerical Schuhmann model for experiments with bed voidage 0.41, (external) interfacial heat transfer using effective pellet conductivity by Beer ($\lambda_{eff} = 0.6$ W/mK)

<table>
<thead>
<tr>
<th>$T_0$ (K)</th>
<th>$T_{final}$ (K)</th>
<th>$T_{avg}$ (K)</th>
<th>flow (Nm³/hr)</th>
<th>$h_{int}$ (W/m²K)</th>
<th>$h_{int}^t$ (W/m²K)</th>
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</table>

Results of packed bed heat transport experiments 113
Errata

page 19, 2nd paragraph; 'equation 2.6-2.7' should be 'equation 2.8-2.9'

page 27, 1st paragraph; 'figure 2.17' should be 'figure 2.18'
  'figure 2.18' should be 'figure 2.19'
  3rd paragraph; 'equation 2.9' should be 'equation 2.11'

page 28, 1st paragraph; 'equation 2.11' should be 'equation 2.17'
  'equation 2.1' should be 'equation 2.5'
  3rd paragraph; 'figure 2.12' should be 'figure 2.13'

page 29, 1st paragraph; 'figure 2.11' should be 'figure 2.12'
  'equation 2.18-2.21' should be 'equation 2.20-2.23'
  'equation 2.32' should be 'equation 2.34'
  'equation 2.36' should be 'equation 2.38'
  'equation 2.26' should be 'equation 2.28'

page 32, 1st paragraph; 'figure 2.11' should be 'figure 2.12'
  'equation 2.35' should be 'equation 2.37'

page 33, 5th paragraph; 'equation 2.37' should be 'equation 2.39'

page 35, 2nd paragraph; 'figure 2.19' should be 'figure 2.27'