DNS of turbulent particle-laden channel flow with heat and mass transfer

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DNS OF TURBULENT PARTICLE-LADEN CHANNEL FLOW
WITH HEAT AND MASS TRANSFER

PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Technische Universiteit
Eindhoven, op gezag van de rector magnificus, prof.dr.ir. C.J. van Duijn,
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door

Emanuele Russo

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“Du täuscht dich, Goldmund. Du nimmst eben an, daß dieser fleißige Rechner immer neue Schulaufgaben löse, die ein Lehrer ihm stellt. Er kann sich die Fragen aber auch selbst stellen, sie können als zwingende Gewalten in ihm entstehen. Man muß manchen wirklichen und manchen fiktiven Raum mathematisch berechnet und gemessen haben, ehe man als Denker an das Problem des Raumes sich wagen kann.”

“You are wrong, Goldmund. You assume that this zealous problem-solver continuously solves problems a teacher poses for him. But he can also ask himself questions; they can arise within him as compelling forces. A man must have measured and puzzled over much real and much fictitious space mathematically before he can risk facing the problem of space itself.”

(Hermann Hesse, *Narziß und Goldmund*)
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Summary

Particle-laden flows are formed by a fluid carrier phase and a solid or liquid dispersed phase. In case of a turbulent flow with a large number of particles whose sizes is smaller than the Kolmogorov length, it is common practice to adopt a point-particle approach. In this thesis we include exchange of momentum, heat and mass between the carrier phase and dispersed phase in an Euler/Lagrangian framework. Only a few studies have been reported on the modeling of heat and mass transfer in a numerical model of turbulent particle-laden flow.

In the first part of the thesis (chapter 2) the carrier phase consists of air and water vapor, while the dispersed phase consists of water droplets which can evaporate. In the second part of the thesis ( chapters 3-5), the dispersed phase consists of biomass and coal particles.

Heat and mass transfer play a crucial role in a channel flow with evaporating droplets. We adopt two-way coupling of heat and mass to analyze the effects of phase change on the global heat transfer properties of the channel and on droplet and water vapor motion. In the water-droplet turbulent channel flow simulations we perform DNS of inhomogeneous turbulent channel flow in which one of the walls is heated while the other is cooled. This leads to non-uniform temperature and vapor mass fraction profiles. We adopt an incompressible formulation for the carrier phase, using a spectral code which was developed in our department and the discrete phase was added to the code for simulating evaporation of the droplets and condensation of the water vapor. In an earlier study it was found that the presence of a large number of inertial particles gives a significant increase in the effective heat transfer between the channel walls. In the present study we investigated to what extent phase changes affect heat transfer. The latent heat of evaporation associated to phase change of droplets yields an enhancement of the heat transfer through the channel by approximately a factor 2.6 compared to solid particles with the same properties as water. The magnitude of the enhancement depends on the initial temperature of the air-vapor mixture in the channel but it is almost independent of the heat flux applied to the walls. Evaporation and condensation lead to growth of droplets near the cold wall and shrinkage near the warm wall. This results in a gradient in water vapor concentration. In the resulting statistically steady state, diffusion and turbulent transport of water vapor lead to a mean flux of water vapor from the warm to the cold wall. Conservation of water mass shows that this must be compensated by an opposite mass flux of water droplets, which is quantified computing the water droplet mass flux. This is the first time
this net flux of water droplets has been directly computed by means of a numerical simulation.

In the second part of the thesis a point-particle model for the pyrolysis of biomass particles is developed, which is coupled to a direct numerical simulation of the turbulent flow of gas in a channel with hot walls, using two-way coupling of mass, momentum and energy. This is the first attempt to model pyrolysis of biomass in a 3D flow framework. Our Euler-Lagrangian formulation offers the advantage to directly simulate the interaction between gas and particles and to accurately predict particle dispersion. These two characteristics are very important in simulating phenomena that strongly depend on gas-particle interaction and particle dispersion. In case of co-pyrolysis, accurate prediction of the number of biomass particles in a certain region around a coal particle is fundamental. These advantages are also the motivation for developing our Euler-Lagrangian formulation to simulate co-firing in the future.

The point-particle model of the biomass pyrolysis is derived from the simplified biomass pyrolysis model proposed by Haseli. First, the model is tested in Matlab assuming that the gas properties are constant. The problem of the stiffness of some equations is solved with approximations which we demonstrate to be sufficiently accurate. In this way the model is made suitable for the same integration method as for the gas phase. The biomass pyrolysis model is implemented in a compressible finite volume code with two-way coupling. We used biomass particle size distributions relevant for practical applications, and we modeled them using a Rosin-Rammler distribution function. The same type of distribution is used to model the coal particle size distributions. However, coal particles have a diameter smaller than the biomass particles. For this reason the simulations with very small coal particles require some more attention to reduce the computational time.

We do not model combustion but focus on pyrolysis, in particular on the effect of particle-gas interaction on the conversion time, i.e. the time needed to convert biomass into char. An accurate prediction of pyrolysis is very important to model and simulate the following biomass combustion, while for coal pyrolysis is generally a negligible step in the modeling. Gas-particle interaction affects the conversion time, which shows a characteristic dependence on particle size and concentration. The conversion time increases with particle concentration but at the same particle concentration, the two-way coupling effect is stronger in the presence of smaller particles, due to the higher total surface area of heat exchange. These two dependencies are analyzed more in detail during co-pyrolysis, showing their effect when varying the blending ratio of the mixture.
Contents

Summary v

1 Introduction 1

2 Water droplet condensation and evaporation in turbulent channel flow 5
  2.1 Introduction ................................................. 5
  2.2 Governing equations ........................................... 8
    2.2.1 Gas .................................................. 8
    2.2.2 Droplets ............................................. 11
    2.2.3 Coupling terms ......................................... 13
  2.3 Numerical method and set up of the simulations .................. 14
  2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer ............................................. 17
    2.4.1 Thermal properties ..................................... 18
    2.4.2 Nusselt number ......................................... 24
    2.4.3 Droplet size distribution ................................ 30
    2.4.4 Droplet migration in the statistically steady state ....... 34
  2.5 Turbulence modulation ........................................ 36
  2.6 Discussion and Conclusions .................................... 39

3 Biomass particle pyrolysis model 41
  3.1 Introduction ................................................ 41
  3.2 Single biomass particle model ................................ 43
    3.2.1 Heating of virgin biomass ................................ 45
    3.2.2 Pre-pyrolysis ......................................... 47
    3.2.3 Pyrolysis ............................................. 48
    3.2.4 Post-pyrolysis heating ................................ 51
    3.2.5 Thermally thick particle ............................... 51
  3.3 Stiffness problem of the equations in the pyrolysis phase ....... 53
    3.3.1 Early stiff phase of pyrolysis ......................... 53
    3.3.2 Thick particle: early stiff phase ...................... 55
    3.3.3 Thick particle pyrolysis with moving thermal front .... 56
    3.3.4 Late stiff phase of pyrolysis ......................... 56
Introduction

Particle-laden turbulent flows can be found in many engineering and natural processes, such as the formation of aerosols, internal combustion in engines and dispersion of pollution in the atmosphere or in oceans [10]. In the field of multi-phase flows, particle-laden turbulent flow remains a challenging topic due to the dynamic nature of particle motion (dispersed phase) which modulates the turbulent characteristics of the flow (carrier phase) through two-way coupling. This means that the particles are affected by the gas but also that the gas is influenced by the particles. Despite the enormous progress in computing power, it is still impossible to simulate millions of particles interacting with a turbulent flow up to all details of the flow. The computing cost for simulating the flow around each individual particle is prohibitive if a large number of particles is present [14, 15, 54].

For some applications in which the particle size is smaller than the Kolmogorov scale, the computing costs can be reduced by adopting a point-particle approach [13, 33]. The particle is treated as a point with concentrated mass and correlations are used for computing the forces acting on the particle, for instance the drag force. The gas phase is solved within an Eulerian framework, while the dispersed phase is solved using Lagrangian tracking of the particles. This approach has been widely used in direct numerical simulation (DNS) and large eddy simulation (LES) [40, 16, 53, 4]. This thesis focuses on an extension of this formulation, which has been less extensively addressed: the effect of mass and heat exchange between carrier phase and dispersed phase.

In this thesis, we present how we model and implement heat and mass exchange of biomass particles undergoing pyrolysis in a turbulent flow of gas. Pyrolysis is thermal degradation (devolatization) in the absence of an externally supplied oxidizing agent. For this application, mass and heat exchange are very important for predicting the behavior of the flow and particles. The detailed numerical simulation of biomass pyrolysis is crucial when predicting the subsequent steps taking place in a power plant, for instance combustion. In this work we present the first attempt to model biomass pyrolysis within a 3-D framework using an approach that describes the interaction between gas and particles in terms of momentum, mass and heat exchange in detail. Moreover, the biomass pyrolysis model we present accounts for moving thermal and pyrolysis fronts and temperature gradients inside the particle, whereas the point-particle approach has traditionally been used for particles with a very small Biot number, i.e. particles with uniform temperature.
The word biomass refers to all kind of materials derived from living or recently living organisms, mostly plant matter and its derivatives, such as wood, wood-derived fuels, fuel crops, agricultural and agro-industrial by-products, and animal by-products. Biomass has attracted the attention of researchers and industrial companies because it is believed to play an important role in the future. Unlike other renewable energies, it is available in large quantities in many regions of the planet. Secondly, but of equal importance, biomass research is important in view of increasing concerns about the environmental impact of most other methods of energy conversion. This work is part of the Clean Combustion Concepts project, which supports industry in the development and application of combustion technologies aiming at stable conversion processes of contemporary and future fuels with optimal efficiency and minimum pollutant emissions. Regarding biomass, the target of reducing CO\textsubscript{2} emissions from coal-fired power plants can be accomplished with co-firing at high biomass fraction and high oxygen concentration (Oxyfuel conditions). Co-firing of coal and biomass reduces greenhouse gas emissions by replacing a certain percentage of coal (fossil fuel) with biomass (renewable fuel). In The Netherlands the target of 50% co-firing in coal-fired power plants is estimated to be reached by 2020. Oxyfuel conditions significantly reduce flue gases and the resulting flue gases have a much higher carbon-dioxide concentration suitable for sequestration.

The work presented in this thesis is part of a larger project named BiOxyFuel supported by the Dutch Technology Foundation STW. The objective of the BiOxyFuel project is to increase understanding and predictive capabilities of torrefied biomass combustion at high co-firing percentages under oxy-fuel conditions. The tasks in this project are divided among three Ph.D candidates. Experimental research is conducted by Eyerusalem Gucho at the University of Twente and focuses on the improvement of biomass properties by means of torrefaction. Single particle modeling was carried out by Yousef Haseli in the Combustion Technology group at Eindhoven University of Technology. The objective of his research was to develop a model for single biomass particles with reduced degrees of freedom that can be used in the particle-laden model presented in this thesis.

A biomass pyrolysis model, which is suitable for DNS using the point-particle approach, must meet certain requirements. In the same way as a semi-empirical cor-
relation for computing the drag force acting on the particle simplifies the problem of accurately solving the flow around a particle, the biomass pyrolysis model must describe the pyrolysis process occurring inside the biomass particle with only a few degrees of freedom. One of the parameters must be the surface temperature of the particle because the particle-gas interaction takes place at the surface and the calculation of the heat transfer between particle and gas requires the surface temperature to be known. The biomass pyrolysis model we propose is described in chapters 3 and 4, along with detailed information about its implementation in a DNS code with two-way coupling.

In chapter 5 DNS of co-pyrolysis of coal and biomass is presented. In general, the pyrolysis and combustion properties of biomass and coal differ significantly. In particular, coal particles are typically smaller than biomass particles and have a higher heating value. In this chapter torrefied biomass is used since torrefied biomass particles are more similar in size to coal than raw biomass particles. The analysis of co-pyrolysis for varying blending ratios gives insight on the effects it has on the pyrolysis process. A higher blending ratio means a higher total volume fraction and a bigger size of the particles. These two properties have counteracting two-way coupling effects on the mass loss history of coal and biomass. Recently, synergetic effects in co-pyrolysis were reported. This effect concerns the interaction between hydrogen gas released during pyrolysis of the biomass particles and the char of coal particles. We developed a first crude model for synergy and studied its effects on the co-pyrolysis results.

The motivation to develop an Euler-Lagrangian formulation for biomass pyrolysis comes from the fact that this process strongly depends on the interaction between the particle and the flow as well as on particle dispersion. However, there are other situations in which this is important. One example is water-droplet laden turbulent flow. In order to investigate the effects of heat and mass transfer on turbulent flow without the extra complication of the additional degrees of freedom present in biomass particles undergoing pyrolysis, we first studied heat and mass transfer between water droplets in turbulent flow of humid air. This is presented in chapter 2, where we study this flow in a channel.
Figure 1.2. Water droplets in air.

Water droplets grow and shrink through evaporation and condensation of water vapor if they are present in air at variable temperature. Therefore, two-way coupling of heat and mass plays an important role. Although the momentum coupling is accounted for, the most interesting effects are a result of heat and mass transfer. In particular, the phase change of the dispersed phase directly affects the heat transfer in a channel. Moreover, it leads to a net flux of water droplets from the cold to the warm wall, which is observed for the first time in a DNS.
Water droplet condensation and evaporation in turbulent channel flow

2.1 Introduction

Particle-laden turbulent flows can be found in many engineering and natural processes, such as the formation of aerosols, internal combustion in engines and dispersion of pollution in the atmosphere or in oceans [10]. In the field of multi-phase flows, particle-laden turbulent flow remains a challenging topic due to the dynamic nature of particle motion (dispersed phase) which modulates the turbulent characteristics of the flow (carrier phase) through two-way coupling. This means that the particles are affected by the gas but also that the gas is influenced by the particles. In this chapter we focus on differentially heated turbulent channel flow in which the dispersed water droplets have a two-way coupling to the gas flow incorporating next to momentum transfer also heat and mass transfer. The consequences for the overall heat transfer, in terms of the Nusselt number, and for the mass transfer, in terms of the size distribution of the droplets, are studied in detail.

The rapid increase in computing power in the past fifty years stimulated research into particle-laden turbulent flows. Particle-laden flows are formed by a fluid phase and a solid or liquid phase. The fluid phase is the “carrier phase” and the solid or liquid phase is the “dispersed phase” [37]. The presence of the dispersed phase requires that the flow around each particle is represented. The most fundamental approach involves full resolution of the flow around each particle. Despite the enormous progress in computing power, it is still impossible to simulate millions of particles interacting with a turbulent flow up to all details of the flow. The computing cost for simulating the flow around each individual particle is prohibitive if a large number of particles is present [14, 15, 54]. Therefore, in case of turbulent flow with large numbers of particles with sizes smaller than the Kolmogorov scale $\eta$, it is common practice to adopt a point-particle approach to keep the computational cost at acceptable levels [13, 33]. In this approximation the motion of a particle is described by the Maxey-Riley equation,
Water droplet condensation and evaporation in turbulent channel flow

which is based on the well-understood physics of low-Reynolds-number flow around a small sphere [39, 49, 5]. This approach allows numerical simulations with millions of particles [12] and has been used to perform direct numerical simulations (DNS) as well as large-eddy simulations (LES) in many applications [40, 16, 53, 4].

The coupling between the phases can be classified into three types: 1) “one-way coupling” if the carrier phase influences the dispersed phase, but the number of particles and the mass loading are too small to have an effect on the carrier phase; 2) “two-way coupling” if the dispersed phase has also a noticeable feedback on the flow; 3) “four-way coupling” when the particle concentration is so large that direct particle-particle interaction is important as well. In the simulations presented in this chapter we adopt two-way coupling, characterized by exchange of momentum, heat and mass with the carrier phase. Generally, for the momentum exchange one must account for the drag force, the body force, the added mass force, the lift force, the pressure drag and the Basset history force. However, the problem can be simplified in case of a water-air system. According to Armenio and Fiorotto [3], for particles with mass density large compared to the fluid mass density, all forces except the Stokes drag force and gravity are negligible.

For point-particle models, the effect of mass and heat transfer in turbulent flows has hardly been addressed. Up to now, only a few studies have been reported on the modeling of heat and mass transfer in a point-particle model that is suitable for numerical computations. In particular, Mashayek [34] was the first to study evaporating droplet dispersion in homogeneous turbulence with two-way coupling of heat and mass transfer, as well as momentum exchange between droplets and fluid. Miller and Bellan [42] performed a complete two-way coupled DNS of a temporally developing mixing layer with one stream laden with evaporating hydrocarbon droplets. Later, this approach was extended to a reacting mixing layer [41] and to study the effect of two-way coupling in a spray including evaporation and combustion phenomena [36, 47, 56]. In a recent work, Masi et al. [38] used this approach to generate data for modelling two-phase flows with evaporation using a statistical approach (Mesoscopic eulerian formalism).

In the present study, we extend the work of Mashayek [34] and consider wall-bounded turbulent flow with evaporation and condensation using DNS. We consider a channel flow at a frictional Reynolds number of 150 in which one of the walls is heated while the other is being cooled. This leads to a gradient of temperature in the wall-normal direction of the channel and hence also to a non-uniform mean vapor mass fraction. We use a similar point-particle approach for the droplets as Mashayek [34], but we adopt an incompressible formulation for the carrier phase and restrict to cases where changes in mass density of the carrier phase due to evaporation and condensation are limited and consistent with a divergence-free velocity field. The governing equations for the point droplets are based on conservation laws and empirical correlations for heat, mass and momentum exchange with the carrier phase.

Moreover, droplet-droplet collisions are ignored because of the small volume fraction considered in our simulations [13]. In the present model, not all mechanisms present in reality have been incorporated. Droplets can grow unrealistically large and nucleation of droplets is not taken into account. In reality large droplets will break up
2.1 Introduction

if their diameter exceeds a critical value which depends on the local shear rate [27, 58].

The effect of these limitations on the results is very small. In the simulations presented here, the largest droplet is still only 35% larger than the initial diameter. Moreover, the relative humidity only slightly exceeds 100%, so that nucleation does not play a role in the cases considered here.

The aim of this study is to analyze the effects of phase change on the global heat transfer properties of channel flow and on droplet size distribution and motion. In power electronics and microelectronics applications, phase change in microchannels was shown to improve the heat exchange due to the high latent heat of evaporation [31]. Another way to improve the heat transfer is employing nanofluids, i.e. a fluid containing nanometer-sized particles. In an earlier study [29] this result was extended to inertial particles in wall-bounded flow. It was found that the presence of a large number of inertial particles gives a significant increase in the effective heat transfer between the channel walls. The particles studied there had the properties of water droplets, but evaporation and condensation were not taken into account. In the present study it will be investigated to what extent phase changes affect the heat transfer. This will be quantified by the Nusselt number, which is the non-dimensional ratio of the temperature gradient at the walls and the average temperature gradient over the height of the channel. The Eulerian-Lagrangian model we adopt allows to analyze the effect of heat transport by droplet transport combined with phase change in wall-bounded flow. The change in Nusselt number can be quantified not only in terms of the change in thermal resistance due to the presence of a dispersed phase but also by the dynamic effect of droplets undergoing phase change transported in the channel and the heat transport associated with them. In order to distinguish between the different contributions to the Nusselt number, additional simulations with solid particles instead of droplets and without particles altogether will be included in the analysis. Moreover, the influence of the mean gas temperature in the channel and the magnitude of the applied heat flux will be studied.

The simulations are started from a homogeneous droplet distribution and a relative humidity of the carries gas of 100%. Initially, the droplets will migrate toward the walls of the channel due to turbophoresis, creating an increase in droplet concentration in the vicinity of the walls. Simultaneously, evaporation and condensation lead to growth of droplets near the cold wall and shrinkage near the warm wall. This results also in a gradient in water vapor concentration, directed from the cold to the warm wall. In the resulting statistically steady state the droplet concentration and mean droplet size are approximately constant. In that situation diffusion and turbulent dispersion of water vapor will lead to a mean flux of water vapor from the warm to the cold wall. Conservation of water mass shows that this must be compensated by a net mass flux of water droplets from the cold to the warm wall. We will analyze the time needed before this equilibrium state is reached and quantify the magnitude of these mass fluxes. To the best of our knowledge this is the first time that such a water cycle has been studied in a numerical simulation.

The chapter is organized as follows. In Section 2 we introduce the governing equations for the air-water vapor carrier phase and the dispersed droplet phase. Section 3 is devoted to a description of the numerical method and the specifications of the
simulations presented in this chapter. In Section 4 we will present and analyze the results of the simulations and in particular address the effects of the evaporating and condensing droplets on the heat transfer between the walls of the channel and the inhomogeneity of droplet size in the wall-normal direction of the channel. In Section 5 the turbulence modulation by evaporating droplets is analysed. Finally, Section 6 is devoted to concluding remarks.

2.2 Governing equations

In this section governing equations for the carrier phase and for the droplets are presented, including the models for momentum, heat and mass transfer between the two phases. We consider a mixture of air and water vapor and refer to this carrier phase as the ‘gas’ in the sequel, while the dispersed phase consists of water droplets. Henceforth, the terms droplet and particle will refer to water droplet and solid particle, respectively. A particle does not exchange mass with the carrier phase, but only energy and momentum. We will first present the equations for the gas phase and then those for the droplets. Since the equations for the gas phase contain coupling terms which depend on the properties of the droplets, we will postpone the specification of these coupling terms to the final part of this section.

2.2.1 Gas

The gas is considered in an Eulerian way and is assumed to behave as an incompressible fluid. This means that the gas mass density is constant, \( \rho_g = \text{const} \). However, it does not mean that the mass densities of air \( (\rho_a) \) and water vapor \( (\rho_v) \) cannot vary. Indeed, both are functions of position \( x \) and time \( t \), \( \rho_g = \rho_a(x,t) + \rho_v(x,t) = \text{const} \).

In the problems we investigate the incompressibility assumption is justified since:
- the Mach number is much smaller than 1.
- the change in mass density due to changes in temperature is in the order of 2% or less.
- in all simulations the gas is initially saturated, with 100% relative humidity everywhere. When the simulation starts the channel is differentially heated in such a way that the amount of water vapor condensing in the colder part of the channel is balanced by evaporation of droplets in the warmer part.

In view of the assumed incompressibility, during the transient phase \( \rho_a \) and \( \rho_v \) vary in time and space, but their sum remains constant, which means that local variations of air mass density are balanced by local variations of water mass density and the same for local variations in time:

\[
\nabla \rho_a = -\nabla \rho_v \quad \text{and} \quad \frac{\partial \rho_a}{\partial t} = -\frac{\partial \rho_v}{\partial t}.
\]

The model for the motion of the gas consists of the continuity equation for incompressible flow, which implies a divergence free flow:

\[
\nabla \cdot \mathbf{u} = 0
\]
and the Navier-Stokes equations for momentum conservation. We adopt the Navier-Stokes equation in rotational form:

\[
\frac{\partial \mathbf{u}}{\partial t} + \omega \times \mathbf{u} + \nabla P = \nu \Delta \mathbf{u} + \frac{\mathbf{F}}{\rho_g} + \frac{L_{\mathbf{u}}}{\rho_g}
\] (2.3)

following Kuerten [28]. In (2.3) \( \omega = \nabla \times \mathbf{u} \) is the vorticity, \( P = p/\rho_g + \frac{1}{2}u^2 \), \( \nu \) is the kinematic viscosity of the gas, \( p \) the static pressure, \( \rho_g \) is the gas mass density, and \( \mathbf{F} \) is the driving force necessary to maintain a constant total mass flow rate. Finally, \( L_{\mathbf{u}} \) is the term describing the momentum exchange between the two phases, that will be specified in section 2.2.3.

The water vapor is also treated in an Eulerian way. The rate of change of the vapor mass density is expressed as the sum of transport due to diffusion, convection, and mass transfer between the two phases by evaporation and condensation:

\[
\frac{\partial \rho_v}{\partial t} + \nabla \cdot (\rho_v \mathbf{u}) - \nabla \cdot (D \nabla \rho_v) = L_v.
\] (2.4)

The term \( L_v \) represents the mass transfer between the droplets and the water vapor, that will be specified in section 2.2.3. We are dealing with a binary system of air and water vapor for which a single diffusion coefficient \( D \) is adequate to represent the diffusion of one species into the other [7, page 502]. This diffusion coefficient is assumed to be constant in space and time, which is motivated by the fact that the solute (water vapor) is present at much lower concentration than the solvent (air). Hence, under all simulated conditions the diffusion of water vapor in dry air will determine the diffusion coefficient \( D \). Moreover, the variations in temperature in all simulations are sufficiently small to use a constant value of \( D \).

The temperature equation is obtained from the equation expressing conservation of internal energy of the two phases. The total internal energy \( E \) in a volume \( \Delta V \) can be written as the sum of the integral of the internal energy density of the gas phase over the volume and the internal energy of all droplets contained in the volume:

\[
E = \int_{\Delta V} \left[ \rho_a c_{v,a} + \rho_v c_{v,v} \right] T_g + \rho_v \ell_0 \, dV + \sum_{i=1}^{N_\Delta} E_i,
\] (2.5)

where \( \rho_a \) (\( \rho_v \)) and \( c_{v,a} \) (\( c_{v,v} \)) are the mass density and specific heat capacity at constant volume of the air (vapor) respectively, \( \ell_0 \) is the latent heat of water at \( T = 0^\circ C \) and \( T_g \) is the gas temperature. Moreover, \( N_\Delta \) is the number of droplets present within \( \Delta V \) and \( E_i \) is the internal energy of droplet \( i \). The specific heat capacities are assumed to be independent of temperature. This is an accurate assumption in the temperature range considered in this chapter, since the specific heat capacities of water and air vary less than 0.1% in the temperature range of each simulation.

In this work we will disregard the contribution of the kinetic energy to the total energy and the work performed by the driving force and by the viscous force. This is justified since the contribution from kinetic energy is at the typical bulk velocity of 1 m/s we consider very small compared to the internal energy. If all kinetic energy in the system would be converted into internal energy, the increase in temperature
would be only approximately $5 \times 10^{-4} \, ^\circ C$. Conservation of energy is incorporated by equating the rate of change of the energy $E$ in $\Delta V$ to the change in energy through the boundaries of the volume due to convection and due to heat conduction and diffusion of vapor. After application of Gauss’s divergence theorem this leads to:

$$\frac{\partial}{\partial t} \left[ (\rho_a c_{v,a} + \rho_v c_{v,v}) T_g + \rho_v \ell_0 \right] + \nabla \cdot \left[ \left( (\rho_a c_{v,a} + \rho_v c_{v,v}) T_g + \rho_v \ell_0 \right) \mathbf{u} \right] = \nabla \cdot \left( k_g \nabla T_g \right) + \nabla \cdot \left( \left( (c_{p,v} - c_{p,a}) (T_g - T_{\ell_0}) + \ell_0 \right) D \nabla \rho_v \right) + L_E. \quad (2.6)$$

The terms on the left-hand side represent the rate of change of internal energy and the convection of internal energy. The first term on the left-hand side is the change in internal energy by heat conduction, where we denote the heat conductivity coefficient of the gas by $k_g$. The second term on the right-hand side represents diffusion arising from variation in composition, which is the dominant diffusion mechanism in case of a water-air system under not too extreme conditions in terms of temperature and pressure (a review is given in Bird et al. [7]). Pressure and temperature gradients are sufficiently small to ignore pressure diffusion and thermal diffusion in this chapter [7, page 566]. The quantities $c_{p,a}$ and $c_{p,v}$ denote the specific heat capacities of air and vapor at constant pressure. The quantity $(T_g - T_{\ell_0})$ is the difference between the gas temperature and the temperature at which the latent heat is evaluated (273.15 K).

The last term on the right-hand side describes the internal energy transfer between the two phases and will be specified below.

Next, substitution of (2.4) and use of (2.1) and (2.2) results in the following governing equation for the gas temperature:

$$\left( \rho_a c_{v,a} + \rho_v c_{v,v} \right) \left( \frac{\partial T_g}{\partial t} + \nabla \cdot (\mathbf{u} T_g) \right) = k_g \nabla^2 T_g + L_{\text{wd}} + L_{\text{diff}} + L_{\text{2way}} \quad (2.7)$$

Although individually $k_g$ and $(\rho_a c_{v,a} + \rho_v c_{v,v})$ are functions of the binary mixture composition, we assume that the heat diffusivity $k_g/ (\rho_a c_{v,a} + \rho_v c_{v,v})$ in (2.7) is constant. For the small values of the vapor mass fraction considered here this assumption appears to be accurate [55]. Note, however, that the dependence of $k_g$ on $\rho_v$ is taken into account in the term $L_{\text{wd}}$ in (2.7), which represents the transport of energy due to diffusion of water vapor and due to the dependence of the thermal conductivity of the gas on the vapor mass density:

$$L_{\text{wd}} = \left\{ \frac{\partial k_g}{\partial \rho_v} + D (c_{p,v} - c_{p,a}) \right\} \nabla \rho_v \cdot \nabla T_g \quad (2.8)$$

Here we used $\nabla k_g = (\partial k_g/\partial \rho_v) \nabla \rho_v$. The third term on the right-hand side is the contribution to the temperature change from diffusion of vapor and is equal to:

$$L_{\text{diff}} = (R_a - R_v) D (T_g - T_{\ell_0}) \nabla^2 \rho_v, \quad (2.9)$$

where $R_a = c_{p,a} - c_{v,a}$ is the specific gas constant of air and similarly $R_v = c_{p,v} - c_{v,v}$ is the specific gas constant of vapor. Finally,

$$L_{\text{2way}} = L_E - ((c_{v,v} - c_{v,a}) T_g + \ell_0) L_v \quad (2.10)$$
represents the contribution from the coupling between the two phases, which will be specified in section 2.2.3.

The time evolution of the gas velocity, gas pressure, gas temperature and water vapor mass density can be calculated from (2.2), (2.3), (2.7) and (2.4). The two-way coupling terms in these equations will be specified after the presentation of the governing equations for the droplets.

### 2.2.2 Droplets

In this subsection the equations of motion, mass, and temperature of the droplets are presented. The droplets are considered in a Lagrangian way. The point-particle approach is used, implying that we also assume the droplets to have uniform temperature. The ratio of the heat transfer resistances inside and at the surface of a droplet can be quantified by the Biot number, which is defined as

\[ Bi = \frac{h_m R}{k_l} \]

with \( h_m \) the convective heat transfer coefficient from gas to droplet, \( R \) the typical radius of a droplet and \( k_l \) the thermal conductivity of the droplet. For the small droplets we consider here the convective heat transfer coefficient equals \( h_m = k_g / R \) with \( k_g \) the thermal conductivity of the carrier gas. Therefore, the typical value of the Biot number is equal to the ratio of the thermal conductivities of water and the carrier gas: \( Bi = 0.046 \). This indicates that the typical time scale of heat conduction inside a droplet is more than 20 times larger than the typical time scale for heat transfer from the carrier gas to a droplet. Therefore, the assumption of uniform droplet temperature is justified.

Since droplets are small and have a much higher mass density than the gas, the drag force is the dominant force exerted by the gas on a droplet [3]. We do not take gravity into account, but it may be added in the spanwise direction to reproduce a real test case. However, settling of droplets is not a focal point in this study. Therefore, Newton’s law applied to a droplet can be written as:

\[
\frac{d(m_i v_i)}{dt} = m_i (\mathbf{u}(\mathbf{x}_i, t) - v_i) \left(1 + 0.15 \frac{Re_p^{0.687}}{\tau_p} \right) + \mathbf{v}(\mathbf{x}_i, t) \frac{dm_i}{dt}. \tag{2.11}
\]

Here, \( m_i \) is the mass of particle \( i \), \( v_i \) its velocity and \( \tau_d \) the droplet relaxation time given by \( \tau_d = \rho_i d_i^2 / (18 \mu_g) \). Moreover, \( Re_p = | \mathbf{v}_i - \mathbf{u}(\mathbf{x}_i, t) | ^2 d_i / \nu \) is the Reynolds number based on the droplet diameter \( d_i \) and the relative velocity between the droplet and the carrier gas at the droplet position, \( \rho_l \) is the mass density of liquid water and \( \mu_g \) the dynamic viscosity of the gas. The two terms on the right-hand side are the drag force, where the standard Schiller-Naumann drag correlation valid for droplet Reynolds numbers between 0 and 1000 is adopted [8], and the change in momentum due to phase change. Equation (2.11) can also be written in terms of accelerations, yielding the following droplet equation of motion:

\[
\frac{d\mathbf{v}_i}{dt} = (\mathbf{u}(\mathbf{x}_i, t) - \mathbf{v}_i) \left(1 + 0.15 \frac{Re_p^{0.687}}{\tau_p} \right). \tag{2.12}
\]

The Lagrangian droplet tracking requires integrating the trajectory equation:

\[
\frac{d\mathbf{x}_i(t)}{dt} = \mathbf{v}_i, \tag{2.13}
\]
in which the particle velocity $v_i$ is computed from (2.12).

In order to derive an equation for the droplet temperature which takes into account the heat and mass transfer to and from the gas, we consider the rate of change of the internal energy of a droplet:

$$\frac{dE_i}{dt} = \frac{d}{dt} (m_i c_l T_i) = h_v \frac{dm_i}{dt} + h_m A_i (T_g(x_i,t) - T_i).$$

(2.14)

This equation states that the variation of the internal energy of droplet $i$ is due to contributions from condensation and evaporation which change the mass of the droplet, and due to heat exchanged by convection at the droplet surface. The heat exchange at the droplet surface is modeled using the heat transfer coefficient $h_m$ as defined by Bird et al. [7]. In a single droplet context, the relevant temperature difference for convection is that between the surroundings and the droplet surface temperature. In (2.14) the surface temperature is the same as the droplet temperature because we assume the droplet temperature to be homogeneous. The temperature of the surroundings is taken as the temperature of the gas $T_g(x_i,t)$ at the location of droplet $i$. Finally, $A_i$ is the surface area of droplet $i$. Kinetic energy is not included here because it is much smaller than the internal energy: the kinetic energy of a droplet with a velocity on the order of 1 m/s is less than 0.05% of its internal energy. In the first term on the right-hand side $h_v$ denotes the specific enthalpy of water vapor. Denoting the specific enthalpy of water as $h_l$, and the droplet volume by $V_i$, (2.14) can be rewritten as:

$$\rho_l c_l V_i \frac{dT_i}{dt} = (h_v - h_l) \frac{dm_i}{dt} + h_m A_i (T_g(x_i,t) - T_i).$$

(2.15)

Here, we used $h_l = c_l T$ and chose the zero point of enthalpy at $T = 0^\circ C$ in the liquid water phase: $h_l(T = 0^\circ C) = 0$. Therefore, the water vapor enthalpy is:

$$h_v = h_v(T = 0^\circ C) + c_{p,v} T_i = h_l(T = 0^\circ C) + \ell_0 + c_{p,v} T_i = \ell_0 + c_{p,v} T_i.$$  

(2.16)

For closing equation (2.15) we need to express $h_m$ and $\frac{dm_i}{dt}$ in terms of known quantities. We use correlations from literature that are applicable to spherical droplets in the range of Reynolds and Prandtl numbers relevant in our simulations. For forced convection around a sphere, the heat-transfer correlation is chosen as [7]:

$$\frac{h_m d_i}{k_g} = 2 + 0.6 \frac{Re_p^{1/2}}{Pr^{1/3}},$$

(2.17)

where $Pr$ is the Prandtl number of the carrier gas.

For the mass transfer we also follow Bird et al. [7], who considered condensation of a hot vapor on a cold surface in the presence of a non-condensable gas. This is applicable in the situation we consider here, since the carrier gas consists of a mixture of water vapor, which is condensable, and the non-condensable air. The resulting expression for the mass transfer is:

$$\frac{dm_i}{dt} = - m_i Sh \frac{1}{3 \tau_p Sc} \ln \left( \frac{1 - x_{v,\delta}}{1 - x_{v,0}} \right),$$

(2.18)
2.2 Governing equations

where the Schmidt number \( Sc = \mu g / (\rho g D) \) and \( x_{v,0} \) are the vapor mass fractions \( x_v = \rho_v / \rho_g \) in the surroundings of the droplet and at the surface of the droplet, respectively. The Sherwood number is given by \( Sh = 2 + 0.6Re_d^{1/2}Sc^{1/3} [7] \).

The driving quantity in (2.18) is the difference \( x_{v,\delta} - x_{v,0} \). Since interfacial resistance is negligible at the droplet surface, we can assume that the air at the droplet surface is saturated [45, 50]. Therefore, with the ideal gas law \( x_{v,0} \) can be written in terms of the particle temperature \( T_i \) and the saturation pressure \( p_{v,sat} \) on the droplet surface as follows:

\[
x_{v,0} = \frac{\rho_{v,sat}}{\rho_g} = \frac{p_{v,sat}}{R_v T_i \rho_g},
\]

(2.19)

where \( R_v \) is the specific gas constant of water vapor. The saturation pressure is calculated using Antoine’s relation [1]:

\[
\frac{p_{v,sat}(T_i)}{p_{ref}} = \exp \left( A - \frac{B}{C + T_i} \right)
\]

(2.20)

where \( T_i \) is in degrees Celsius, \( p_{ref} \) is the reference pressure of \( 10^5 \text{ Pa} \), and the coefficients are: \( A = 11.6834 \), \( B = 3816.44 \degree \text{C} \), and \( C = 226.87 \degree \text{C} \). For \( x_{v,\delta} \) we take the vapor mass fraction of the gas at the position of the droplet, \( \rho_v(x_i, t) \).

The time evolution of particle velocity, position, temperature and mass can be calculated from equations (2.12), (2.13), (2.15) and (2.18). In the next section the two-way coupling terms between the gas phase and the droplets will be specified.

2.2.3 Coupling terms

The two-way coupling terms in the governing equations for the gas phase satisfy the requirement that these terms do not change the total water mass, total momentum and total internal energy in the system. They only transfer mass, momentum and internal energy from one phase to the other. Moreover, we will assume that the two-way coupling terms act as a point force in the governing equations for the gas.

Based on these two considerations the two-way coupling term in the Navier-Stokes equation for the gas phase (2.3) can be written as:

\[
L_u = - \sum_{i=1}^{N} \frac{dm_i}{dt} v_i \delta (x - x_i) = - \sum_{i=1}^{N} m_i \frac{dv_i}{dt} \delta (x - x_i) - \sum_{i=1}^{N} v_i \frac{dm_i}{dt} \delta (x - x_i).
\]

(2.21)

The coupling term is split into two contributions representing the momentum transfer from the droplet to the gas due to acceleration and due to mass transfer arising from evaporation or condensation respectively. The delta-function \( \delta (x - x_i) \), indicates that the coupling terms act only at the locations of the droplets.

The two-way coupling term in the water vapor equation (2.4) follows from conservation of water mass and is given by:

\[
L_v = - \sum_{i=1}^{N} \frac{dm_i}{dt} \delta (x - x_i).
\]

(2.22)
The two-way coupling term in the gas temperature equation (2.7) follows from conservation of internal energy and using (2.15) for the change in particle temperature as:

\[
L_{\text{2way}} = - \sum_{i=1}^{N} \left( c_{p,v}T_i + (c_{v,a} - c_{v,v})T_g \right) \frac{dm_i}{dt} \delta(x - x_i) - \sum_{i=1}^{N} h_m A_i (T_g - T_i) \delta(x - x_i) = L_{\text{evap}} + L_{\text{conv}}
\]

where \(L_{\text{evap}}\) and \(L_{\text{conv}}\) represent the gain (loss) of energy for the carrier phase due to evaporation (condensation) of the droplets, and the convective heat transfer between fluid and droplets, respectively.

In the next section the numerical method will be described, including the way in which the coupling terms are evaluated in the numerical method.

### 2.3 Numerical method and set up of the simulations

In this section we first sketch the main elements of the numerical approach and pay special attention to the coupling between the discrete droplet phase and the continuous gas phase. Next, the geometry of the channel and the applied boundary conditions are described. Then, the parameters and initial conditions used in the simulations are presented.

The proposed model for two-way coupling in droplet-laden turbulent flow is applied in DNS of turbulent channel flow. First, the numerical method adopted for the continuous phase is described, next the one for the droplets. Finally, the method for treating the coupling terms numerically is discussed.

DNS of turbulent channel flow is performed using an extended version of the pseudo-spectral code as described in Kuerten [28]. The extensions consist of the incorporation of equations for the water vapor mass density and for the temperature and of two-way coupling of momentum, vapor mass and internal energy between the gas and the droplet phase. The application of periodic boundary conditions in the streamwise and spanwise directions allows the use of a Fourier-Galerkin approach, whereas in the wall-normal direction a Chebyshev-collocation method is used. The time integration method for the gas consists of a combination of a third-order accurate compact-storage explicit Runge-Kutta method for the nonlinear terms (including the coupling terms) and the implicit Cranck-Nicolson method for the viscous and pressure terms. The nonlinear terms are computed in physical space using fast Fourier transform and the 3/2-rule to prevent aliasing errors. The velocity field is divergence free within machine accuracy by applying the influence matrix method. For further details concerning this spectral method for the gas phase see Kuerten [28].

The time integration of the gas phase requires that the coupling terms are computed at the same times as the other terms in the governing equations of the gas phase. Since, apart from a minus sign, the same coupling terms are needed in the equations for the droplets as in the equations for the gas phase, the same Runge-Kutta method is applied to the governing equations of the droplets. The gas properties (velocity, temperature, and vapor mass density) at the droplet location are calculated
2.3 Numerical method and set up of the simulations

by second-order accurate tri-linear interpolation. It is known from literature that a fourth-order interpolation in DNS leads to negligible differences in the statistical particle properties [33].

The coupling terms of each droplet are computed at the droplet location and successively distributed to the eight surrounding mesh points using the same weights as used for the tri-linear interpolation of the gas properties to the droplet positions. This means that in this numerical formulation the delta function \( \delta (x - x_i) \) is implemented in such a way that the transfer is spread over the eight grid points neighboring the droplet. Subsequently, the two-way coupling terms resulting from the contributions of all droplets are added to the nonlinear terms in the gas equations in physical space and transformed to Fourier space. In case only a few droplets are present, this way of calculating the coupling terms may lead to numerical oscillations. However, the contributions to high wave numbers caused by the delta function are for a large part canceled if the number of droplets is of the same order as the number of grid points, as in the simulations reported here [29].

The domain has a size of \( 4\pi H \) in streamwise direction and \( 2\pi H \) in spanwise direction, where \( H \) is half the channel height (see figures 2.3). For the streamwise, wall-normal, and spanwise directions the notation \( x, y, \) and \( z \) is used, respectively. In both homogeneous directions (spanwise and streamwise) 128 Fourier modes are used, and 129 Chebyshev collocation points are employed in the wall-normal direction. The grid points are clustered near the walls in order to resolve the boundary layer. Periodic boundary conditions are applied in the streamwise and spanwise directions and the no-slip condition is adopted at the walls. On the upper (warm) wall a uniform and constant heat flux is supplied to the channel, which is equal to the heat flux extracted from the lower (cold) wall in order to conserve the total thermal energy of the system. For the droplets, periodic conditions are applied in the homogeneous flow directions, which means that when a droplet leaves the domain in one of these directions, it re-enters the domain from the other side with the same properties. If a droplet approaches a wall within a distance of its radius, rules of elastic collision of a hard spherical particle are applied without heat transfer.

Simulations are performed at frictional Reynolds number approximately \( Re_\tau = 150 \), based on friction velocity \( u_\tau = \sqrt{\tau_w/\rho_g} \) and half the channel height, where \( \tau_w = \mu g \frac{\partial \langle u \rangle}{\partial y} \bigg|_{y=\pm H} \) is the wall shear stress and \( \langle u \rangle \) the mean streamwise velocity component, averaged over the two homogeneous directions and time. Marchioli et al. [33] also show that the flow at this Reynolds number has reached a state of fully developed turbulence. The gas parameters are computed using a mass-density weighted average according to the dry air and water vapor concentrations. The droplets or solid particles have an initial diameter \( d_i/H = 3.09 \times 10^{-3} \) which corresponds to a Stokes number \( St = \tau_p^+ = \tau_p u_\tau^2/\nu = 10 \). This initial droplets size ensures that during the simulation the size of growing particles near the cold wall does not exceed the Kolmogorov length. On the other hand, smaller particles would results in an attenuated turbophoresis effect, especially in the warm part of the channel where evaporation occurs. There are two million droplets or particles, which are initially randomly and homogeneously distributed over the channel domain. The initial volume fraction is approximately \( 2.2 \times 10^{-4} \) which is in the range where droplet-droplet collisions can
be ignored but two-way coupling is relevant [13]. During the simulation particles are clustered near the walls due to turbophoresis. Here, the two-way coupling formulation remains still valid although the local particle concentration is very close to the upper bound of the two-way coupling regime. The initial droplet velocity and temperature are taken the same as the gas velocity and temperature at the droplet location.

The flow is initialized by a turbulent velocity field in the statistically steady state of fully developed turbulence obtained from a simulation without droplets at the same Reynolds and Prandtl number. The initial temperature is homogeneous and when the simulation starts, a constant heat flux is imposed at both walls to gradually generate a temperature gradient in the wall-normal direction. The magnitude of the heat flux is shown in table 2.1. On the upper wall, the heat flux is positive, while it is negative on the lower wall. The initial relative humidity \( \phi \), which is the ratio of the partial water vapor pressure and the saturation pressure, equals 100% all over the channel. Hence, the air-vapor mixture is in saturation conditions. It is noted that this relative humidity equals \( \rho_v T_g(x)/\rho_{v,sat} T_i(x) \) and, apart from the initial condition, is therefore not equal to the normal relative humidity which is assessed at identical temperatures: \( \rho_v T_g(x)/\rho_{v,sat} T_g(x) \). In the initial transient phase the temperature changes because of the applied thermal boundary conditions. Consequently, the saturation conditions change according to the local change of the temperature. The applied heat fluxes lead to evaporation of droplets in the upper half of the channel (warmer part), while condensation will occur in the lower half (colder part). Using these settings we simulate an experiment in which an initially isothermal channel flow with droplets is heated at the top wall and cooled at the bottom wall.

We investigated five test cases as described in Table 2.1. In the reference test case the initial temperature is 20°C, hereafter we refer this case as ‘reference’. The case named ‘solid’ is identical to ‘reference’ but the droplets are replaced by solid particles,
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

<table>
<thead>
<tr>
<th>Case</th>
<th>Particles</th>
<th>Initial temperature (°C)</th>
<th>heat flux (W/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>reference</td>
<td>water droplets</td>
<td>20</td>
<td>32</td>
</tr>
<tr>
<td>high temperature</td>
<td>water droplets</td>
<td>50</td>
<td>34</td>
</tr>
<tr>
<td>high heat flux solid</td>
<td>water droplets</td>
<td>20</td>
<td>160</td>
</tr>
<tr>
<td>solid</td>
<td>solid particles</td>
<td>20</td>
<td>32</td>
</tr>
<tr>
<td>no particles</td>
<td></td>
<td>20</td>
<td>32</td>
</tr>
</tbody>
</table>

Table 2.1. Definition of the test cases.

so that phase transition between the gas and particle phases is not possible. Case ‘no particles’ uses the same settings for the boundary conditions and flow parameters, but is without particles. The other two cases named ‘high temperature’ and ‘high heat flux’ serve to investigate the effects of different physical conditions. In ‘high temperature’ the initial temperature is increased from 20°C to 50°C, while in ‘high heat flux’ the heat flux applied at the walls is five times higher than in the reference case.

2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

In this section the simulation results of the test cases will be presented and analyzed. In test cases with water droplets several phenomena play a role. The heat flux applied at both walls leads to a non-uniform temperature profile in the wall-normal direction. Near the warm wall droplets will evaporate and near the cold wall water vapor will condense. In this way a non-uniform water vapor concentration will arise, which leads on average to transport of vapor from the warm to the cold wall. On a larger time scale the inhomogeneity of the turbulence in the wall-normal direction leads to turbophoresis through which droplets will move on average from the center of the channel toward the walls, where the temperature and hence the mean droplet size attain different values. This, together with the turbulent motion of droplets will lead to a gradually increasing variance of droplet size. In this section we will first study the thermal properties of the flow and in particular focus on the heat transfer between the walls quantified by the Nusselt number. Next, we will consider the droplet size and investigate the mean transport of water including both droplets and vapor.

In the analysis presented in this section we make use of quantities averaged over the homogeneous directions. This averaging is denoted by bars over the corresponding variable. When considering the statistically steady state we additionally average over time. Quantities averaged over the two homogeneous directions and time are denoted by brackets, $\langle \cdot \rangle$. We distinguish between gas quantities and quantities of the dispersed phase. The gas quantities are known in all Chebyshev collocation points and averaging over homogeneous directions can easily be performed both in Fourier space and in real space (Eulerian averaging). Quantities of the dispersed phase are only known at the locations of the droplets or particles. Averages over homogeneous directions of the dispersed phase are computed by averaging over all droplets that at
some time are contained in a slab parallel to the channel walls (Lagrangian averaging). Specifically, we divide the channel into uniformly spaced bins in the wall-normal direction and compute the average profiles using these bins. We use 40 bins, since that number is sufficiently large to accurately represent the dependence of the statistical quantities on the wall-normal coordinate and sufficiently small to minimize statistical errors arising from a too small number of particles in one bin. For the computation of statistical properties of the two-way coupling terms in the gas equations and the relative temperature between particles and carrier gas, the bins defined by the Chebyshev collocation points are used in order to obtain these quantities in the same points as the other statistical properties of the carrier gas. It should be noted that due to their size no particles can be present in the first two grid cells neighboring both walls. Therefore, two-way coupling terms equal zero there. Since droplets can have an arbitrary size, small droplets could be present in these cells, but in the simulations reported here this has not been observed.

2.4.1 Thermal properties

We consider a channel flow in which we impose a constant heat flux at the walls. This creates a temperature gradient in the wall-normal direction, which leads to evaporation (condensation) in warmer (colder) regions, since initially the air is saturated. In figures 2.2 the fluid temperature of the reference case is shown, averaged over the homogeneous directions, as a function of the normalized wall-normal coordinate $y/H$ at several instances in time ($t^+$ is the dimensionless time $t^+ = t u_2^2/\nu$). The figure shows the development of an inhomogeneous temperature profile from the uniform initial temperature. A steady temperature profile is reached around $t^+ = 5,000$, after which the mean temperature exhibits only small fluctuations around the steady profile. As a consequence of evaporation and condensation induced by the changing temperature, the mean water vapor mass fraction changes in time as figures 2.3 shows. An inhomogeneous water vapor concentration profile in the wall-normal direction is seen to develop, which closely resembles the mean temperature profile. Also for the water vapor concentration a steady profile is reached around $t^+ = 5,000$.

In the statistically steady state, the difference in temperature between the channel walls depends considerably on whether there are particles present in the flow or not, and whether the particles can undergo phase change or not. This is illustrated by a comparison of the reference case (with water droplets) with the case with solid particles and with the case without particles. The initial and boundary conditions are the same for the three cases and the properties of the solid particles are the same as those of the water droplets with the exception that no evaporation or condensation can occur. Figure 2.4 shows the mean temperature profiles for these three cases as functions of the wall-normal coordinate in the statistically steady state. It can be seen that the presence of particles has an influence on the heat transfer properties of the channel, which is in agreement with the results shown in Kuerten et al. [29]. The temperature difference between the walls reduces considerably in case solid particles are added to a clean single-phase flow. Adding droplets instead of solid particles gives rise to an even larger decrease in the temperature difference between the walls.

In order to understand the reason for the reduced temperature difference in case
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.2. Reference case: mean gas temperature as a function of the wall-normal coordinate at several instances of time - $t^+ = 0$ (•••••); $t^+ = 400$ (——); $t^+ = 800$ (—); averaged over a long time interval in the statistically steady state for temperature, $5,000 < t^+ < 16,000$ (---).

Figure 2.3. Reference case: vapor mass density normalized by its uniform initial value as a function of the wall-normal coordinate at several instances of time - $t^+ = 0$ (•••••); $t^+ = 400$ (——); $t^+ = 800$ (—); averaged over a long time interval in the statistically steady state for vapor mass density, $5,000 < t^+ < 16,000$ (---).
droplets are added instead of particles we focus on the region close to the upper, warm wall. Similar arguments apply to the lower, cold wall. We will first identify the terms in the equation for the temperature of the carrier gas that play an important role in the near-wall region. Then we will explain the sign of these terms and compare their magnitude for the reference, solid, and no particles cases. Finally, we will use this to understand the difference in temperature evolution between the three cases.

Figure 2.5 shows the temperature histories at the warm wall for the three cases: reference, solid, and no particles. The different temperature response seen in the three cases starts in the early stages of the transient phase. Since at $t^+ \approx 30$ the difference between solid and no particles is well visible, we analyze the terms in the energy equation (2.7) at this time. Figure 2.6 shows the magnitude of the terms in the energy equation, averaged over the homogeneous directions, in the region close to the warm wall for the reference case. We distinguish the gas convection term $\left( - (\rho_a \ c_{v,a} + \rho_v \ c_{v,v}) \nabla \cdot (u \ T_g) \right)$, the thermal diffusion term $k_g \nabla^2 T_g$, the two-way coupling term due to phase change $L_{\text{evap}}$, the two-way coupling term due to convective heat transfer $L_{\text{conv}}$ and the terms due to diffusion of water vapor ($L_{\text{wd}} + L_{\text{diff}}$). The figure shows that the two-way coupling term due to evaporation and the terms due to diffusion of water vapor are negligible and that the gas convection term is small compared to the remaining two terms. In the transient phase, the thermal diffusion term is not fully balanced by the sum of the gas convection term and the two-way coupling term, which results in an increasing gas temperature in the region close to the upper wall.
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.5. Mean temperature history of the warm wall - reference case with water droplets (---), solid particles (--.--); no particles (--.-).

Figure 2.6. Terms in the energy equations for the reference case as a function of the wall-normal coordinate close to the warm wall at $t^+ = 32$: diffusion term $k_g \nabla^2 T_g$ (---), gas convection term $-(\rho_u c_{v,a} + \rho_v c_{v,v}) \nabla \cdot (u T_g)$ (--.--), convective flux coupling term $\mathcal{L}_{\text{conv}}$ (---), evaporation coupling terms $\mathcal{L}_{\text{evap}}$ (-----), $\mathcal{L}_{wd} + \mathcal{L}_{\text{diff}}$ (••••).
As can be expected from the shape of the temperature profile, the thermal diffusion term tends to increase the gas temperature. Since the particles can only be heated by heat transfer from the gas through two-way coupling, their temperature is lower than the gas temperature near the warm wall. Therefore, the two-way coupling term tends to decrease the gas temperature. The gas convection term results in a small decrease of the gas temperature.

Now we know which terms play a role in the change of the gas temperature in time, we compare the non-negligible terms, i.e. the gas convection and diffusive terms and the two-way coupling term due to convective heat transfer between the two phases, for the reference, solid and no particles cases in figure 2.7. This figure shows that the gas convection term is small for all three cases. The absence of the two-way coupling term in the case without particles explains why the temperature increase in this case is larger than in the other two cases. In the no particles case the thermal diffusion term is smaller than in the reference and solid cases. However, since the two-way coupling term is absent in the no particles case, the sum of all terms in the gas temperature equation is larger than in the reference and solid cases and hence the increase of the gas temperature is larger. The difference in temperature between the reference case with water droplets and the case with solid particles is mainly caused by the two-way coupling term due to convective heat transfer. This term is larger in magnitude in the reference case than in the solid case, because the evaporation in the reference case leads to a lower temperature of the dispersed phase. The resulting larger temperature difference between droplets and carrier gas increases the magnitude of the two-way coupling term.

The thermal properties of the air-water system depend on the initial temperature and via the heat and mass transfer process on the strength of the thermal forcing through the walls. This can be observed by comparing the high temperature case and high heat flux case with the reference case. The results of these two cases show some differences with respect to the reference case. In particular, the asymmetry in the mean gas temperature profile of the high heat flux case shown in Fig. 2.8 is due to the convex dependence of the saturation pressure on temperature expressed by Antoine’s relation (2.20). The asymmetry of the temperature profile is not clearly visible in the reference and high temperature cases because of the small temperature difference between the two walls in the statistically steady state. Apparently, a higher overall temperature does not lead to strongly different normalized temperature profiles. When the temperature difference between the walls becomes larger, also the asymmetry of the temperature profile becomes visible. This is reflected in the water vapor mass concentration as well. Apart from this, the results of the high heat flux case are very similar to the reference case. A larger difference in the heat transfer properties of the channel occurs in high temperature case, i.e. for a higher initial temperature, and this will be analyzed in the next section.
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.7. Terms in the energy equations as a function of the wall-normal coordinate close to the warm wall at $t^+ = 32$ - upper lines are the diffusive fluxes, middle lines the advection fluxes and lower lines the convection coupling terms; reference case (---), case with solid particles (--.--), case without particles (-.-).

Figure 2.8. High heat flux case: mean gas temperature as a function of the wall-normal coordinate in the statistically steady state - temperature in the upper half of the channel (---), mirror temperature in the bottom half part of the channel (--.--).
2.4.2 Nusselt number

In order to quantify the heat transfer properties in the statistically steady state we introduce the non-dimensional Nusselt number, defined as:

\[ \text{Nu} = \left( \frac{d\langle T_g \rangle |_{\text{wall}}}{\langle \Delta T_g \rangle} \right) \frac{\Delta T_g}{2H}, \]

where \( \Delta T_g \) is the mean temperature difference between the walls. A larger Nusselt number implies that the fluid is able to transfer more heat between the walls at the same wall temperature difference. Such a process is better suited in a heat exchanger.

In the transient regime we define the Nusselt number as a function of time without averaging in time:

\[ \text{Nu}(t) = \left( \frac{dT_g}{dy} |_{\text{wall}} \right) \frac{\Delta T_g}{2H}. \]

In figure 2.9 Nu\((t)\) is shown for the five cases. As already explained in the previous subsection, we observe that the presence of particles or droplets leads to a larger value of the Nusselt number in the statistically steady state (compare reference, solid and no particles cases). Moreover, Nu depends on the initial channel temperature (compare reference and high temperature cases), but hardly on the applied heat flux at the walls (compare reference and high heat flux cases).

Following Kuerten et al. [29] we quantify the effect of particles and droplets on Nu by splitting it into various contributions. Averaging equation (2.7) over the homogeneous directions and over time in the statistically steady state and integrating the result twice over the wall-normal direction, we obtain the following expression for Nu:

\[
\text{Nu} = 1 - \frac{1}{\alpha_g \langle \Delta T_g \rangle} \int_{-H}^{H} \langle u'_y T'_g \rangle dy + \frac{1}{\langle \Delta T_g \rangle} \int_{-H}^{H} \int_{-H}^{H} \left( \frac{L_{\text{wd}} + L_{\text{diff}} + L_{\text{2way}}}{k_g} \right) ds dy, \tag{2.24}
\]

where \( \alpha_g = k_g / (\rho_a c_{v,a} + \rho_v c_{v,v}) \) is the thermal diffusivity, which is assumed constant in this work. Moreover, \( u'_y = u_y - \langle u_y \rangle \) is the fluctuating wall-normal component of the velocity of the gas, \( T'_g = T_g - \langle T_g \rangle \) is the fluctuating gas temperature and \( \langle \Delta T_g \rangle \) is the mean temperature difference between the two walls in the statistically steady state. In (2.24) the contributions from the water vapor diffusion terms \( L_{\text{wd}} \) and \( L_{\text{diff}} \) and from the two-way coupling term due to evaporation \( L_{\text{evap}} \) appear to be negligible, in agreement with the results shown in figure 2.6. For all cases considered they contribute less than 0.1% to Nu.

The contribution from \( L_{\text{diff}} \) can be analyzed in more detail by writing it as:

\[
(R_a - R_v) D \nabla (T_g - T_{\ell_0}) \nabla^2 \rho_v = (R_a - R_v) D \nabla \rho_v \cdot \nabla (T_g - T_{\ell_0}) + (R_v - R_a) D \nabla \cdot (\nabla (T_g - T_{\ell_0}) \nabla \rho_v). \tag{2.25}
\]

The first term on the right-hand side is of the same form and magnitude as \( L_{\text{wd}} \). The second term can be integrated if the thermal conductivity is assumed constant...
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.9. Nusselt number history for the five cases - reference case ( ), high temperature case ( ), high heat flux case ( ), solid particles ( ), no particles ( ). The inset zooms in on the initial behavior.

to give a contribution to Nu equal to

$$\text{Nu}_{\text{diff}} = \frac{(R_v - R_a)D}{k_g \langle \Delta T_g \rangle} \int_{-H}^{H} (T_g - T_{\ell_0}) \frac{\partial \rho_v}{\partial y} dy.$$  \hspace{1cm} (2.26)

In order to analyze this term further we write \(T_g - T_{\ell_0} = T_0 + T''_g - T_{\ell_0}\), where \(T_0\) is the constant initial temperature. Since \(T''_g\) is negative in the lower half of the channel and positive in the other half, while \(\frac{\partial \rho_v}{\partial y}\) is positive throughout the channel, it can be expected that the term with \(T''_g\) in \(\text{Nu}_{\text{diff}}\) is very small. The numerical analysis of this term shows that it is three orders of magnitude smaller than \(\text{Nu}_{\text{diff}}\). The contribution from \((T_0 - T_{\ell_0})\) can be integrated over \(y\), which results in:

$$\text{Nu}_{\text{diff}} \approx \frac{(R_v - R_a)D(T_0 - T_{\ell_0})\langle \Delta \rho_v \rangle}{k_g \langle \Delta T_g \rangle},$$  \hspace{1cm} (2.27)

where \(\langle \Delta \rho_v \rangle\) is the mean vapor mass density difference between the two walls in the statistically steady state. For the small temperature differences between the walls in our simulations, we can write

$$\langle \Delta \rho_g \rangle \approx \frac{\partial \rho_{v,\text{sat}}}{\partial T} \langle \Delta T_g \rangle,$$  \hspace{1cm} (2.28)

since deviations from saturation are very small in the statistically steady state. The result is:

$$\text{Nu}_{\text{diff}} \approx \frac{\partial \rho_{v,\text{sat}} (R_v - R_a)D(T_0 - T_{\ell_0})}{k_g \partial T}.$$  \hspace{1cm} (2.29)
Table 2.2. Different contributions to the Nusselt number for all test cases.

<table>
<thead>
<tr>
<th>Case</th>
<th>Particles</th>
<th>Nu</th>
<th>Nu_{lam}</th>
<th>Nu_{turb}</th>
<th>Nu_{part}</th>
</tr>
</thead>
<tbody>
<tr>
<td>reference</td>
<td>water droplets</td>
<td>15.6</td>
<td>1.0</td>
<td>3.0</td>
<td>11.6</td>
</tr>
<tr>
<td>high temperature</td>
<td>water droplets</td>
<td>35.9</td>
<td>1.0</td>
<td>2.5</td>
<td>32.4</td>
</tr>
<tr>
<td>high heat flux</td>
<td>water droplets</td>
<td>15.6</td>
<td>1.0</td>
<td>3.0</td>
<td>11.6</td>
</tr>
<tr>
<td>solid</td>
<td>solid particles</td>
<td>5.9</td>
<td>1.0</td>
<td>3.4</td>
<td>1.5</td>
</tr>
<tr>
<td>no particles</td>
<td>-</td>
<td>4.4</td>
<td>1.0</td>
<td>3.4</td>
<td>-</td>
</tr>
</tbody>
</table>

For the reference case and the high heat flux case we find \( \text{Nu}_{\text{diff}} \approx 0.003 \) and for the high temperature case the result is \( \text{Nu}_{\text{diff}} \approx 0.03 \), which shows that this contribution to the Nusselt number is negligible indeed.

Hence, the Nusselt number can accurately be expressed as \( \text{Nu} = \text{Nu}_{\text{lam}} + \text{Nu}_{\text{turb}} + \text{Nu}_{\text{part}} \), where \( \text{Nu}_{\text{lam}} = 1 \) is the result for laminar flow without particles,

\[
\text{Nu}_{\text{turb}} = -\frac{1}{\alpha_g \langle \Delta T_g \rangle} \int_{-H}^{H} \langle u'_y T'_g \rangle dy
\]

is the contribution from turbulent transport and

\[
\text{Nu}_{\text{part}} = \frac{1}{\langle \Delta T_g \rangle} \int_{-H}^{H} \int_{-H}^{y} \frac{\ln \langle T_g(x_i,t) - T_i \rangle}{\langle \Delta T_g \rangle} ds dy
\]

consists of the effect of the convective heat transfer between the two phases.

Table 2.2 shows that the presence of particles or droplets with a two-way coupling to the fluid flow does not significantly reduce the heat transfer due to turbulent transport. This differs from the results obtained by Kuerten et al. [29], and can be explained by the different flow conditions, in particular the much smaller volume fraction of particles in the present simulations. Comparison of reference, solid and no particles cases shows that the presence of particles leads to an increase of the Nusselt number of around 30%. If the particles are replaced by droplets, the increase in Nusselt number is much stronger: a factor of 3.5. For both droplets and particles the increase is caused by the contribution from the convective heat transfer between the two phases, \( \text{Nu}_{\text{part}} \). The simplified expression for \( \text{Nu}_{\text{part}} \) [29],

\[
\text{Nu}_{\text{part}} \approx -\int_{-H}^{+H} \int_{-H}^{y} \frac{2\pi \ln \langle s \rangle d_i \langle T_g(x_i,t) - T_i \rangle}{\langle \Delta T_g \rangle} ds dy,
\]

helps to understand this. The contribution from the convective heat transfer between the phases is large, if the relative temperature is large in regions of the channel with a large concentration of the dispersed phase. Therefore, in the following, we analyze the temperature difference between the two phases and the concentration of particles/droplets.

Figure 2.10 shows the mean temperature difference \( \langle T_g(x_i,t) - T_i \rangle \) averaged over the homogeneous directions near the warm wall at two instances of time for reference and solid cases. Note that \( T_g(x_i,t) \) equals the mean temperature of the gas evaluated at the particle or droplet position. Since particles and droplets are preferentially
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

located at locations where the gas temperature differs from its Eulerian mean, this temperature is not equal to the mean Eulerian gas temperature [29]. The mean temperature difference near the warm wall is initially large for both reference and solid case. At $t^+ = 400$ the mean temperature difference is much smaller for solid particles than with droplets, because a droplet decreases its temperature by evaporation.

In the statistically steady state, which starts later than the times shown in figure 2.10, the mean temperature difference is attenuated by the two-way coupling in both reference and solid cases, resulting in the equilibrium mean relative temperature difference shown in figure 2.11. With solid particles $\langle (T_g(x_i,t) - T_i) \rangle / \Delta T_g$ is larger than with droplets, apart from a small region very close to the walls. The relative temperature difference is multiplied with the particle concentration in expression (2.30) for $\text{Nu}_{\text{part}}$. Figure 2.12 shows that in both cases the majority of the particles is located very close to the walls. The integrand of (2.30) is shown in figure 2.13, where it appears that in the vicinity of the wall the product of particle concentration and relative temperature difference is much larger in the reference case than in the solid case. This is a direct consequence of the phase transition process of water droplets and it results in the observed eight times larger contribution to the Nusselt number from $\text{Nu}_{\text{part}}$.

The test cases with high temperate and high heat flux demonstrate the effect of variations in initial temperature and heat flux imposed at the walls on the heat transfer characteristics. The Nusselt number for the high heat flux case with the same
Figure 2.11. $\langle (T_g(x_i, t) - T_i) \rangle / \langle \Delta T_g \rangle$ as a function of the wall-normal coordinate in the statistically steady state - reference case (---), high temperature case (---), solid particles case (---).

Figure 2.12. Droplet and particle concentration, $n(y)/n_0$ with $n_0$ the initial number of particles in a bin, as a function of the wall-normal coordinate in the statistically steady state - reference case (---), solid particles case (•).
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

initial temperature as the reference case but a five times higher heat flux imposed at the walls is approximately the same as for the reference case. In contrast, the case with high initial temperature results in a much higher Nusselt number. For the following reason the rate of mass transfer near the walls is higher with a higher initial gas temperature. A given temperature change of a drop results in a bigger change in saturation vapor pressure at higher gas temperature. The latter is easily seen from the Clausius-Clapeyron equation or from the Antoine relation (2.20). Initially in the simulations, the gas near the hot wall is heated by heat diffusion from the wall. The drops increase in temperature by convective heat transfer from the gas and evaporation sets in. The higher the initial temperature, the higher the rate of evaporation is for the above reasons. In the statistically steady state, the reference and high temperature cases have about the same drop diameters near the walls but the gas temperature level is higher with a higher initial temperature and hence the mass transfer rate too. The effective heat transfer rate between the two walls is increased by this process and therefore the temperature difference between the two walls is less in the reference case than in the high temperature case, both cases having the same imposed heat flux at the walls. This lower temperature difference causes the higher Nusselt number found. The independence of the Nusselt number on the heat flux imposed at the walls shows that the temperature difference between the two walls in the statistically steady state is proportional to the imposed heat flux.
2.4.3 Droplet size distribution

In this subsection we analyze the change in size of the droplets due to the thermal forcing applied to the channel. In figure 2.14, the mean droplet diameter history normalized by its initial value is plotted at three positions in the channel for the simulation of the reference test case. The three positions are the mid plane of the channel and the first bins near the upper and lower walls. Obviously, near the heated wall the droplets start evaporating, resulting in a decreasing mean diameter, whereas the droplets near the cooled wall grow due to condensation of water vapor. In the middle of the channel the mean droplet diameter fluctuates around its initial value. For $t^+ > 0.5 \times 10^4$ the mean temperature difference between gas and droplets (figure 2.11) and for $t^+ > 2.0 \times 10^4$ the droplet number density (figure 2.15) are constant in time and depend on the wall-normal coordinate. The root-mean-square of the droplet diameter in a certain bin, $d_{rms}$, however, increases at all times, see figure 2.16. If the same droplets would remain in a bin, $d_{rms}$ would not increase. This implies that there is a continuous inflow and outflow of droplets in a bin, as a consequence of turbulent transport. On average, however, the mean drop size history of figure 2.14 appears to follow roughly the following relation [21]:

$$\langle d \rangle^2 = d_0^2 - \beta t,$$

(2.31)

where the so-called evaporation parameter, $\beta$, is a constant. After introduction of a single drop in a flow at different temperature, $T_g$ and certain vapor concentration, $x_{v,0}$, first a transient period occurs and next a steady-state evaporation state sets in. In this steady state, the above relation holds with

$$\beta = 8 \frac{\rho_g D}{\rho_l} \ln \left( \frac{1 - x_{v,\delta}}{1 - x_{v,0}} \right).$$

(2.32)

In the present work the value of $\beta$ will be different, since we consider the droplet size averaged over a distribution in turbulent conditions, where droplet temperature and gas temperature fluctuate in time and space. Moreover, the value of $\beta$ will depend on time, since the relative humidity, which is the driving force for condensation and evaporation, depends on time. Yet, it is remarkable that the above simple relation seems to hold approximately for the mean of a droplet size distribution of a droplet population that is not constant in time. It will come in handy in the interpretation of an asymmetry in the channel flow, below.

Initially, the variance of the droplet diameter in the middle of the channel is smaller than near the walls (figure 2.16). This is caused by the fact that evaporation and condensation only occur close to the walls. The effect of droplet dispersion becomes evident in a later stage. The root-mean square of droplet diameter did not yet saturate at $t^+ = 5 \times 10^4$, the final time of the simulation. Simulating for even longer time, and also including the explicit break-up of droplets that have grown beyond a certain size, will clarify this. In the present work the simulations have been kept sufficiently short to avoid the need to model break-up of droplets.

At the final time of the simulation and close to the wall the root-mean square of the droplet diameter exceeds the change in mean droplet size over time. That implies
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.14. Reference case: Normalized mean droplet diameter history at three locations in the channel ($d_0 = \text{diameter at } t^+ = 0$) - near the warm wall (---), mid plane (-----), near the cold wall (----).

Figure 2.15. Reference case: History of droplet concentration, $n(y)/n_0$ with $n_0$ the initial number of particles in a bin, near the walls - near warm wall (---), near cold wall (----).
that the probability density functions of droplet size of both walls overlap. This is a further illustration of the importance of turbulent transport of droplets through the channel.

Figure 2.17 shows the mean droplet diameter histories near the two walls for the reference, high temperature and high heat flux cases. The higher initial temperature does not affect the droplet mean diameter as much as the higher heat flux. On both walls, the reference and high temperature cases have almost equal mean droplet diameter. In the high heat flux case, evaporation of the droplets near the warm wall and condensation of vapor on the droplets near the cold are enhanced by the higher temperature difference between the walls. In this case, the increase in mean droplet diameter near the cold wall is smaller than the decrease near the warm wall. The main reason for the asymmetry is the different values of the evaporation constant, $\beta$. Figure 2.18 shows the rms of droplet diameter near both walls for the high temperate and high heat flux case. With a higher heat flux the rms near the cold wall is significantly smaller than near the warm wall but much larger than in the high temperature case. For a higher initial temperature, this difference is attenuated since the higher thermal conductivity of the channel leads to a smaller wall temperature difference.

Figure 2.14 shows that the mean droplet diameter has become approximately constant in time after $t^+ = 3.2 \times 10^4$. In this time the mean flow has traveled approximately 250 times the length of the channel. The fact that also the droplet concentration near the wall is approximately constant for $t^+ > 20,000$ (figure 2.15) indicates that an equilibrium is established between condensation and evaporation on the one hand and the water vapor heat flux across the channel on the other hand. We investigate this in more detail in the next subsection.
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.17. Normalized mean droplet diameter history near the cold and warm wall - reference case (—), high temperature case (—), high heat flux case (—).

Figure 2.18. History of the droplet diameter root-mean square - high temperature case near the warm wall (—), high temperature case near the cold wall (—), high heat flux case near the warm wall (—), high heat flux case near the cold wall (—).
2.4.4 Droplet migration in the statistically steady state

There are three mechanisms by which water can have a mean motion in the wall-normal direction of the channel. The first one is molecular diffusion of water vapor by a mean concentration gradient, the second is turbulent transport of water vapor and the third is mean motion of droplets in the wall-normal direction. In the statistically steady state, which is reached at \( t^+ \approx 2.0 \times 10^4 \), the mean droplet size and concentration do not change anymore. Therefore, the three mechanisms should cancel each other.

Since the water vapor concentration is higher at the warm wall than at the cold wall, there will be a net molecular diffusion flux of water vapor from the warm to the cold wall, which at all wall-normal positions in channel is given by \(-Dd\langle \rho_v \rangle / dy\). The direction of the turbulent transport flux of water vapor, \( \langle u'_y \rho'_v \rangle \), can be found in the following way. The water vapor concentration is correlated to the gas temperature (see figures 2.2 and 2.3) and from the fact that \( \text{Nu}_{\text{turb}} \) is positive we know that \( \langle u'_y T^*_g \rangle \) is negative. Therefore, also the flux of water vapor by turbulent transport is negative, i.e. directed from the warm to the cold wall. This implies that, for the three mechanisms to cancel, there must be a net mass flux of water droplets from the cold to the warm wall, which balances the net mass flux of vapor from the warm to the cold wall. We write the total mass flux of water as \( j_{\text{water}} = -Dd\langle \rho_v \rangle / dy + \langle u'_y \rho'_v \rangle + j_{\text{drops}} \), where \( j_{\text{drops}} \) is the mass flux of water droplets, which can be directly calculated in the Lagrangian tracking of droplets adopted in our formulation.

Figure 2.19 shows the mass fluxes for the three mechanisms as functions of the wall-normal coordinate. The results are averaged over the two homogeneous directions and time in the statistically steady state. The calculation of \( j_{\text{drops}} \) is quite critical for two reasons. First, although the droplet concentration near the walls has reached a statistically steady state at \( t^+ \approx 20,000 \), the fluctuations in time are large. A very long time average is required to eliminate the effects of these fluctuations on \( j_{\text{drops}} \). Second, the net mass flux of droplets is small. If only droplets with a positive wall-normal velocity component are considered, the mass flux is larger by two orders of magnitude. Hence, there is a very large ‘traffic’ of droplets to and from the walls separately, balancing almost perfectly and leaving a much smaller \( j_{\text{drops}} \) as result. For these two reasons the mass flux of the droplets is still quite irregular. Still, the results indeed underpin the picture sketched above. Both vapor mass fluxes are directed from the warm to the cold wall and are almost constant when added. The molecular diffusion is dominant in the near-wall regions, whereas the turbulent contribution takes over away from the walls. The net droplet mass flux is directed from the cold to the warm wall. This shows that there is a circulation of water. Water vapor moves from the warm to the cold wall, where it condenses onto droplets. The droplets move from the cold to the warm wall, where they evaporate again.

The circulation model for water in the system is further supported by figure 2.20, which shows the relative humidity \( \phi \) as a function of the wall-normal coordinate at various instances in time. The figure shows that \( \phi \) is very close to 100% in the major part of the channel. Only close to the walls larger deviations from saturation can be seen. In the transient stage these deviations are smaller than 1% and they are even smaller in the statistically steady state. Nevertheless, near the warm wall
2.4 Analysis of the effect of phase transition of droplets on turbulent heat transfer

Figure 2.19. Water vapor and droplet mass fluxes as functions of the wall-normal coordinate in the statistically steady state (averaged over $21,250 < t^+ < 85,300$) in the reference case - water vapor mass flux due to Fick's law ($- - -$), water vapor mass flux due to turbulent diffusion $\langle v_p \rho \rangle$ (-----), droplet mass flux $j_{\text{drops}}$ (- - -).

Figure 2.20. Reference case: relative humidity ($\phi$) history - steady state (averaged over $32,000 < t^+ < 51,000$) (---), $t^+ = 800$ (-----).
\( \phi \) is smaller than 100\%, so that net evaporation will occur and close the cold wall the small supersaturation results in a net condensation of water vapor. If the mean droplet concentration observed in the steady state is constant then the mean net flux of droplets measured by the number of droplets moving from one wall to other one is zero. However, because droplets migrating from the cold wall to the warm wall are bigger in size than droplets migrating in the opposite direction, the mean net mass flux of water droplets cannot be zero and yields the calculated net mass flux.

### 2.5 Turbulence modulation

In this section the effect of phase transition on the turbulence of the channel flow is analyzed. We compare the modulation of turbulence caused by the presence of solid particles or evaporating droplets with the case without particles.

Turbulence modulation by particles is extensively reviewed by [4] and [11], with a remark on the limits of the point-particle method in predicting turbulence modulation when particles are comparable in scale to the small scale turbulence. In our simulations the particle size is smaller than the smallest Kolmogorov scale, also in the near wall region. Figure 2.21 shows the rms of the gas velocity components for the case without particles, the case with solid particles and the reference case with evaporating droplets. The presence of particles leads to a decrease of the turbulent kinetic energy. This result is in agreement with the results of [44] (in the absence of gravity) who reported attenuated turbulent intensities and Reynolds stresses for particles smaller than the dissipative length scale, whereas larger particles augment both intensities and stresses. In the following, the results of test cases with droplets are not shown since there are no significant differences between the turbulence modulation by solid particles and evaporating droplets, also for different initial temperature or boundary conditions, as in the high temperature case or high heat flux case. This negligible effect of phase transition is explained in the following analysis of the turbulence kinetic energy.
Figure 2.21. Rms of the streamwise (A), wall-normal (B), and spanwise (C) velocity components in the steady state - reference case with droplets (…….), with solid particles (----), and without particles (-----).
The turbulence kinetic energy is influenced by the presence of particles, especially in the vicinity of the wall where the particle concentration is higher in the steady state. Two-way coupling of momentum leads to an extra term in the turbulent kinetic energy equation of the gas, associated to $L_u$ in (2.21). In the steady state, the production of turbulent kinetic energy in the gas is balanced by dissipation and particle dissipation due to the drag force between particles and gas, $L_u$:

$$\int_{-1}^{+1} \left( \langle u'_j u'_k \rangle \frac{\partial \langle u \rangle_j}{\partial x_k} \right) dy = \int_{-1}^{+1} \left( \nu \frac{\partial u'_j}{\partial x_k} \frac{\partial u'_j}{\partial x_k} \right) dy + \int_{-1}^{+1} \left( \sum_{i=1}^{N} \frac{d (m_i v_{i,j})}{dt} \delta (x - x_i) \right) dy. $$

(2.33)

Other terms in the TKE equation only transport kinetic energy within the channel and therefore their integrals over the wall-normal direction are equal to zero. Note that the quantity $\langle u \rangle_j$ in the droplet dissipation term is Eulerian averaged, whereas the other quantities are Lagrangian averaged. The quantity $d(m_i v_{i,j})/dt$ can be split in two contributions, in the same way as (2.21). The contribution due to mass transfer arising from evaporation or condensation is negligible since the evaporation rate term is very small, as already shown in the gas temperature budget equation. Other studies, such as [35] and [57], have shown that evaporation leads to an increase in the turbulence kinetic energy of the gas due to the transfer of kinetic energy from particles to gas and attenuation of particle dissipation because of the smaller mass loading ratio due to evaporation. These effects are important at high evaporation rates occurring by starting a simulation from a non-equilibrium state. In our simulation the air-vapor gas is initially in equilibrium. Therefore, during the simulation of the transient phase, the boundary conditions applied result in small evaporation and condensation rates whose effect is negligible.

In order to analyze the dissipative effect of the particles, the integrands of the terms in (2.33) are shown as functions of the channel height in figure 2.22 for the case with solid particles and without particles. The production and dissipation of turbulent kinetic energy decreases due to the presence of particles. Due to the higher particle concentration, this is clearly visible near the walls. The droplet dissipation term in (2.33) is small compared to the other terms. The inset shows that this term produces turbulence kinetic energy near the walls and dissipates it in the core region of the channel. However, its integral is negative and, therefore, it globally has a dissipative effect.

The particle dissipation term is not the only cause for the particle dissipative effect on the gas. In the transient phase, this term is also small compared to the production and the dissipation term. However, due to the turbulence modulation of the particles, the turbulent kinetic energy production is almost immediately attenuated while the
dissipation is still comparable to the case without particles. In addition, the droplet dissipation term adds extra dissipation which yields a decrease in the turbulent kinetic energy which persists until the steady state is reached. The decrease of production is an indirect effect of the particle modulation of the gas velocity field. The mean gas streamwise velocity is accelerated by the presence of particles modulating $\partial \langle u \rangle / \partial y$ and, as already explained in the beginning of this section, also $\langle u' w' \rangle$ is modulated. Of these two contributions to the production term the decrease of the Reynolds stress by the particles is the most important reason for the turbulence attenuation by the presence of particles.

### 2.6 Discussion and Conclusions

In this chapter a numerical simulation method for water droplet-laden turbulent flow including heat and mass transfer by convection and phase change has been discussed. The model has been implemented in a pseudo-spectral DNS code for turbulent channel flow, where the droplets are modeled in a Lagrangian way. The effects of phase change have been studied by comparison with results of a simulation in which only heat transfer between the phases is considered.

The results show that the boundary conditions applied to the initial state leads to evaporation and condensation. After a longer time an equilibrium state is reached in which the relative humidity is close to 100% over the total height of the channel. In the equilibrium state the heat transfer between the two walls, quantified by the Nusselt number, is larger than in case the droplets are absent or replaced by solid
particles with the same physical properties. Moreover, a significant further increase in Nusselt number has been found if the initial temperature of the channel is increased.

Apart from mean temperature, vapor mass density and relative humidity, also the droplet size distribution has been analyzed. The mean droplet diameter is smaller near the warm wall than near the cold wall. When the mean diameter near the walls remains approximately constant in time, an equilibrium between the vapor mass flux and droplet migration is established. The vapor flux from the warm wall to the cold wall is approximately balanced by droplet migration from the cold to the warm wall.

In the simulations that have been performed nucleation of droplets and droplet break up are ignored. Extension of the model with these mechanisms will be a topic of future research.
Biomass particle pyrolysis model

In this chapter a point-particle model for the pyrolysis of biomass particles based on the model developed by Haseli et al. [25] is presented, which will be used in direct numerical simulation of turbulent flow of a gas in a channel with hot walls using two-way coupling of mass, momentum and energy.

The development of the model is described in detail and, in the next chapter, its application in the direct numerical simulation is discussed.

3.1 Introduction

Biomass has attracted the attention of researchers and industrial companies because it is believed to play an important role in the future since, unlike other renewable energies, it is available in large quantities in many regions of our planet [22]. Secondly, but of equal importance, biomass research is important in view of increasing concerns about the environmental impact of most other methods of energy conversion. This work is part of the Clean Combustion Concepts project which supports industry in the development and application of combustion technologies aiming at stable conversion processes of contemporary and future fuels with optimal efficiency and a minimum of pollutant emissions. Regarding biomass, the target of reducing CO₂ emissions from coal-fired power plants can be accomplished with co-firing at high biomass fraction and high oxygen concentration.

The word biomass refers to all kind of materials derived from living, or recently living organisms, mostly plant matter and its derivatives, such as wood, wood-derived fuels, fuel crops, agricultural and agro-industrial by-products, and animal by-products [32]. In this work, we consider both torrefied and non-torrefied wood as biomass. Torrefaction or drying is a thermochemical treatment of biomass at 200 to 320°C. During this process, carried out in the absence of oxygen, the water contained in the biomass is released, obtaining dry biomass. The torrefaction helps to produce much better fuel quality for combustion and gasification applications and improves the grindability and the hydrophobicity, very important for the biomass to be stored outside. The energy required for torrefaction is partly compensated by the energy reduction for grinding torrefied wood compared to raw wood [46]. In chapters 3 and
4 we will consider non-torrefied wood, whereas in chapter 5, where we consider the co-pyrolysis of biomass and coal particles, we use torrefied wood as biomass. In both cases we will adopt the formulations for drag force on a particle and heat transfer between particle and gas for spherical particles. Two of the main differences between wood and torrefied wood are the smaller size and the more spherical shape of the torrefied biomass particle [2]. Therefore, the assumption of spherical particles is a better approximation for torrefied wood than for the raw material. Incorporation of the effects of a non-spherical particle shape on the correlations for drag force and convective heat transfer is a useful topic for further research.

Combustion of biomass involves various physical and chemical processes, which take place successively and even simultaneously [23, 24]. The combustion process can be divided into several general processes: drying/torrefaction, pyrolysis, gasification and combustion. Here, we focus on modeling the pyrolysis process, which can be defined as thermal degradation (devolatization) in the absence of an externally supplied oxidizing agent. During pyrolysis, the virgin biomass is converted into char and volatiles are released. The volatiles consist mainly of tar, carbonaceous charcoal and gases of low molecular weight. In addition, since biomass contains oxygen, considerable amounts of CO and CO$_2$ can be formed during pyrolysis. The volatile composition, concentration and thermal properties depend on the fuel type, temperature, pressure, heating rate and reaction time [32].

The pyrolysis model presented here is based on the model developed by Haseli et al. [25]. Various models of a single biomass particle undergoing pyrolysis are available in literature. These describe the process in terms of partial differential equations since the temperature in a biomass particle depends both on time and on the spatial coordinates. These models are able to capture the variation of important particle quantities during pyrolysis. Haseli presents an overview of the available models in [25]. However, the complexity of these models requires a computational cost which is unfeasible when dealing with a large number of particles undergoing thermo-chemical conversion and interacting with the surrounding turbulent gas flow. In some applications, like the design of combustors or gasifiers, only a few parameters are of interest, for example the mean temperature or surface temperature of the particles, the amount of volatiles released during pyrolysis, the ignition time and the conversion time of the particles. The model proposed by Haseli et al. [25] has the advantage to be suitable in combination with DNS since, based on assumptions, it approximates the solution of the governing partial differential equations by a set of associated ordinary differential equations for the temperature of each particle.

In this chapter, the single biomass particle model of Haseli is presented. Then, the equations of each phase of a biomass particle undergoing pyrolysis are derived. The steps necessary to obtain the ordinary differential equations from the work of Haseli et al. [25] and the subsequent steps to enable the coupling of the model to the DNS of the turbulent gas flow are described. The point-particle model used in the next chapter is based on these equations. In some of the phases of the pyrolysis process the set of governing ordinary differential equations is stiff, which complicates the coupling to the numerical method applied in the DNS. Methods to avoid this and reduce the stiffness of the equations are presented in section 3.3. In section 3.5 a
detailed analysis of the biomass particle pyrolysis model is presented for developing a more efficient numerical method for this problem and study the stability of the biomass pyrolysis equations.

3.2 Single biomass particle model

The biomass pyrolysis model of Haseli et al. [25] consists of several phases for modeling the overall process. In fact, the pyrolysis process is only one of these phases. The other phases take place before and after the pyrolysis phase and do not involve chemical conversion. All the phases are described in detail in this section.

Consider a spherical particle with a uniform initial temperature. When a positive heat flux $\dot{q}$ is applied to its surface, the surface temperature starts to rise and, due to the thermal conductivity of the particle, the temperature profile inside the particle starts to change. Theoretically, the velocity at which the thermal disturbance propagates through the particle is infinite. However, following Haseli et al. [25], immediately after the heat flux is applied, the particle is divided in two regions, the outer region closer to the surface, in which the temperature has changed, and the inner region around the center of the particle, in which the temperature is still unchanged (Figure 3.1 (a)). The surface which separates these two regions is called thermal front and moves from the particle surface towards the particle center. Although the thermal front is assumed to move with a finite velocity, the model has been demonstrated to predict results which agree well with a full thermal model. Hereafter, this phase of the biomass pyrolysis will be called heating of virgin biomass.

When the temperature of the particle center starts to change, the second phase, called pre-pyrolysis heating, starts (Figure 3.1 (b)). The first two phases are different only in the mathematical formulation because in the heating of virgin biomass phase there is a moving thermal front. The pyrolysis phase starts when the surface reaches the pyrolysis temperature. During the pyrolysis phase (Figure 3.1 (c)), the char front moves from the surface towards the center of the particle, since the model is based on the shrinking core model. A further assumption is that the decomposition occurs instantaneously in the thin layer where the temperature equals the pyrolysis temperature. This allows to make use of an integral method, which transforms the partial differential equations into ordinary differential equations. Haseli et al. [25] remark that, although the choice of a fixed pyrolysis temperature may be controversial, according to other studies [18] the process can be well described by assigning a suitable value for this temperature, depending on the process conditions, in such a way that the overall energy balance agrees well with experimental results.

Finally, when the particle is completely converted, the char particle is still heated until it reaches the gas temperature. This phase is called post-pyrolysis heating (Figure 3.1 (d)).
Figure 3.1. Stages of a biomass particle undergoing pyrolysis: heating of virgin biomass (a), pre-pyrolysis (b), pyrolysis (c), post-pyrolysis heating (d)

Figure 3.2. Stages of a thermally thick biomass particle undergoing pyrolysis: heating of virgin biomass (a), thermally thick (e), pyrolysis (c), post-pyrolysis heating (d)
If the particle diameter exceeds a certain value, depending on initial temperature, pyrolysis temperature and heat flux, it may happen that pyrolysis starts before the thermal front has reached the particle center (Figure 3.2 (e)). Therefore, Haseli et al. [25] distinguish thermally thin and thermally thick particles. The phases of a thermally thin and a thermally thick particle are illustrated in figures 3.1 and 3.2, respectively. In the following, first the model for a thermally thin particle will be introduced and then the model for a thermally thick particle will be presented.

### 3.2.1 Heating of virgin biomass

The rate of change of a thermally thin particle during the first phase, the heating of virgin biomass, is described by the following governing equation (in spherical coordinates) and boundary conditions:

\[
\frac{\partial T}{\partial t} = \alpha_B \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) \tag{3.1}
\]

\[
k_B \left. \frac{\partial T}{\partial r} \right|_{r=R} = \dot{q} \tag{3.2}
\]

\[
\left. \frac{\partial T}{\partial r} \right|_{r=r_t} = 0 \tag{3.3}
\]

\[
T(r_t) = T_0. \tag{3.4}
\]

Equation (3.1) describes diffusion of heat inside a spherical biomass particle with constant thermal diffusivity \(\alpha_B\). In this equation \(T\) denotes the temperature, \(r\) the radial coordinate and \(t\) the time. Equation (3.2) is the boundary condition at the particle surface with radius \(R\). The thermal conductivity of the particle equals \(k_B\) and it is constant. At the thermal front, where \(r = r_t\), the temperature and its radial derivative are continuous, as expressed by (3.3) and (3.4), where \(T_0\) is the initial particle temperature. The external heat flux is denoted by \(\dot{q}\) and is the sum of convective and radiative heat flux:

\[
\dot{q} = h(T_g - T_s) + \sigma \epsilon (T_{w}^4 - T_s^4). \tag{3.5}
\]

The convective heat flux is proportional to the relative temperature \((T_g - T_s)\) between the surrounding gas and the particle surface. In this chapter, the gas temperature \(T_g\) is assumed constant, so \(\dot{q}\) only depends on time through its dependence on \(T_s\). In the next chapter, where the particle equations are coupled to the gas equations, the surrounding gas temperature will be time and space dependent \(T_g = T_g(x(t), t)\), so the heat flux will also explicitly depend on time: \(\dot{q} = \dot{q}(t)\). The heat-transfer coefficient \(h\) is computed using correlation (2.17) for a spherical particle. The radiative flux takes into account the heat flux received by the particle from the walls, whose temperature is indicated by \(T_w\). The Stefan-Boltzmann constant and the emissivity are indicated by \(\sigma\) and \(\epsilon\), respectively.

A basic approximation in Haseli’s model is that the temperature profile is parabolic:

\[
T(r) = \phi_2 (R - r)^2 + \phi_1 (R - r) + \phi_0 \quad \text{for} \quad r \geq r_t.
\]
Substitution of the boundary conditions (3.2)–(3.4) yields the coefficients as:

\[
\phi_0 = T_0 + \frac{\dot{q}}{2k_B} (R - r_t)
\]

\[
\phi_1 = -\frac{\dot{q}}{k_b}
\]

\[
\phi_2 = \frac{\dot{q}}{2k_B (R - r)}.
\]

Thus, the biomass temperature profile is:

\[
T(r) = T_0 + \frac{\dot{q}}{2k_B} (r - r_t) \left\{ 1 - \frac{R - r}{R - r_t} \right\} \text{ for } r > r_t. \quad (3.6)
\]

Integration of (3.1) from \( r = r_t \) to \( r = R \) yields:

\[
\frac{d}{dt} \left\{ \int_{r_t}^{R} r^2 T \, dr \right\} = -r_t^2 T_0 \frac{dr_t}{dt} + \int_{r_t}^{R} \alpha_B \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) \, dr
\]

\[
= -r_t^2 T_0 \frac{dr_t}{dt} + \alpha_B \left[ R^2 \frac{\partial T}{\partial r} \bigg|_{r=R} - r_t^2 \frac{\partial T}{\partial r} \bigg|_{r=r_t} \right]
\]

\[
= -r_t^2 T_0 \frac{dr_t}{dt} + \frac{\alpha_B}{k_B} R^2 \dot{q}. \quad (3.7)
\]

On the other hand, substitution of (3.6) in the integral yields:

\[
\frac{d}{dt} \left\{ \int_{r_t}^{R} r^2 T \, dr \right\} = \frac{d}{dt} \left\{ \frac{\dot{q} R^4}{10} - \frac{3}{20} \dot{q} R^3 r_t + \frac{1}{60} \dot{q} R^2 r_t^2 + \frac{1}{60} \dot{q} R r_t^3 + \frac{1}{60} \dot{q} r_t^4 \right\} + \frac{1}{k_B} \frac{dr_t^3}{dt} \frac{1}{3} T_0 \frac{dr_t}{dt} \quad (3.8)
\]

Combining (3.7) and (3.8) it follows that:

\[
\frac{d}{dt} (\dot{q} f(r_t)) = 60 \alpha_B \dot{q} \quad (3.9)
\]

with \( f(r_t) = 6R^2 - \dot{q} R r_t + r_t^2 + \frac{r_t^3}{R} + \frac{r_t^4}{R^2} \). The thermal front position \( r_t \) is expressed in terms of \( T_s \) by evaluating (3.6) at \( r = R \):

\[
r_t = R - \frac{2k_B}{\dot{q}} (T_s - T_0). \quad (3.10)
\]

Equation (3.9) is the governing equation in the first phase. The unknown in (3.9) is \( T_s \). Equation (3.9) can be written in the final form:

\[
\frac{d}{dt} T_s = 60 \alpha_B \dot{q} / \left( f \frac{\partial \dot{q}}{\partial T_s} + \dot{q} \frac{df}{dr_t} \left( \frac{\partial r_t}{\partial \dot{q}} \frac{\partial \dot{q}}{\partial T_s} + \frac{\partial r_t}{\partial T_s} \right) \right). \quad (3.11)
\]
For small values of $t$, $T_s$ will be close to $T_0$ and $\dot{q}$ can be assumed constant. A Taylor expansion of $f(r_t)$ for $T_s$ close to $T_0$ shows that $f(r_t)$ is proportional to $(T_s - T_0)^2$. This is explained more in detail in section 3.5. It follows that $(T_s - T_0) \sim \sqrt{t}$. Therefore, we use a differential equation for $(T_s - T_0)^2$, which equals:

$$\frac{d}{dt} (T_s - T_0)^2 = 120\alpha_B \dot{q} (T_s - T_0) / \left( f \frac{\partial f}{\partial T_s} + \dot{q} \frac{df}{dr} \left( \frac{\partial r_t}{\partial q} \frac{\partial q}{\partial T_s} + \frac{\partial r_t}{\partial T_s} \right) \right).$$  \hspace{1cm} (3.12)

We derive the governing equations in the other phases in the following section.

### 3.2.2 Pre-pyrolysis

This phase is described by (3.1) and the following boundary conditions:

$$k_B \frac{\partial T}{\partial r} \bigg|_{r=R} = \dot{q} \hspace{1cm} \text{(3.13)}$$

$$\frac{\partial T}{\partial r} \bigg|_{r=0} = 0. \hspace{1cm} \text{(3.14)}$$

The difference with the previous phase is the absence of the thermal front. The boundary condition (3.13) represents the heat flux applied at the particle surface and (3.14) is the symmetry boundary condition in the center of the particle. Assuming the temperature profile to be approximated by a parabola:

$$T(r) = \phi_2 (R - r)^2 + \phi_1 (R - r) + \phi_0 \quad \text{for} \quad 0 \leq r \leq R$$

and using the boundary conditions, we obtain the coefficients:

$$\phi_0 = T \bigg|_{r=R} = T_s$$

$$\phi_1 = -\dot{q}/k_B$$

$$\phi_2 = \dot{q}/(2k_BR).$$

Thus, the temperature profile can be written as:

$$T(r) = T_s - \frac{\dot{q}}{k_B} (R - r) + \frac{\dot{q}}{2k_BR} (R - r)^2. \hspace{1cm} (3.15)$$

The rate of change of the integral of $r^2T$ between 0 and $R$ using (3.15) yields:

$$\frac{d}{dt} \left\{ \int_0^R r^2T dr \right\} = \frac{d}{dt} \left\{ \frac{1}{3} R^3T_s - \frac{1}{15} \frac{R^4\dot{q}}{k_B} \right\}. \hspace{1cm}$$

On the other hand, substituting (3.1) in the integral gives:

$$\frac{d}{dt} \left\{ \int_0^R r^2T dr \right\} = \int_0^R \alpha_B \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) dr$$

$$= \alpha_B R^2 \frac{\partial T}{\partial r} \bigg|_{r=R} = \frac{\alpha_B}{k_B} R^2 \dot{q}$$

$$= \alpha_B R^2 \frac{\partial T}{\partial r} \bigg|_{r=R} = \frac{\alpha_B}{k_B} R^2 \dot{q}$$
Requiring these expressions to be equal leads to the governing equation in this phase:

$$\frac{d}{dt} \left( T_s R - \frac{1}{5} R^2 \dot{q} \right) = 3 \frac{\alpha B}{k_B} \dot{q}$$

(3.16)

which is solved for $T_s$ by numerically integrating the following equation:

$$\frac{dT_s}{dt} = 3 \frac{\alpha B}{k_B} \dot{q} / \left( R - \frac{1}{5} R^2 \frac{\partial \dot{q}}{\partial T_s} \right)$$

(3.17)

in which $\frac{\partial \dot{q}}{\partial T_s}$ is obtained by differentiation of (3.5) with respect to $T_s$.

### 3.2.3 Pyrolysis

In the pyrolysis phase, the particle is divided in two regions separated by the interface at which pyrolysis occurs instantaneously. The position of the interface $r_c$ is identified by the char front moving from the surface towards the center of the particle. In this model we assume that the temperature profiles in each region are given by quadratic functions as follows:

$$T(r) = \phi_2 (r-r_c)^2 + \phi_1 (r_c - r) + \phi_0 \quad \text{for} \quad 0 \leq r \leq r_c$$

(3.18)

$$T(r) = \psi_2 (r-r_c)^2 + \psi_1 (r-r_c) + \psi_0 \quad \text{for} \quad r_c \leq r \leq R.$$  

(3.19)

Another model proposed by Haseli in which the temperature profiles are approximated by linear functions predicts also results well in agreement with this model and leads to a more comprehensive model [25].

The heat equations governing this problem are:

$$\frac{\partial T}{\partial t} = \alpha_B \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right)$$

(3.20)

for $0 \leq r \leq r_c$

$$\frac{\partial T}{\partial t} = \alpha_C \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) + (1 - \omega) \frac{m}{4\pi r^2 \alpha_{vol}} \frac{\partial T}{\partial r}$$

(3.21)

for $r_c \leq r \leq R$

with $\alpha = \alpha_C$, the thermal diffusivity of the char. The second term on the right hand side of (3.21) represents heat transfer between the volatiles released at the char front and the char during their transport towards the particle surface. In (3.21) $\alpha_{vol} = C_{vol}/\rho C_c$ and $\omega = \rho C / \rho B$, with $C_{vol}$ and $C_c$ the volatiles and char heat capacities, respectively.

This problem consists of two heat diffusion problems similar to the pre-pyrolysis phase, connected to each other at the interface $r_c$. Because of a moving char front and the co-existence of these two regions, in the pyrolysis phase there are three governing equations, whereas in the other phases there is only one. These equations are derived in the following. For the sake of simplicity, we assume $\omega = 0$ in the derivation of the governing equations. The extra term arising from the volatiles interaction with char is presented in the end without showing its derivation.
The boundary conditions of (3.20) and (3.21) are:

\[
k_C \frac{\partial T}{\partial r} \bigg|_{r=R} = \dot{q} \tag{3.22}
\]

\[
\frac{\partial T}{\partial r} \bigg|_{r=0} = 0 \tag{3.23}
\]

\[
T_B \big|_{r=r_c} = T_C \big|_{r=r_c} = T_p \tag{3.24}
\]

\[
4\pi k_B r_c^2 \frac{\partial T}{\partial r} \bigg|_{r=r_c} = 4\pi k_C r_c^2 \frac{\partial T}{\partial r} \bigg|_{r=r_c} + m_h \Delta h_p. \tag{3.25}
\]

In the boundary condition at the surface of the particle the thermal conductivity of the char \(k_C\) is used. The latter condition at the interface represents the continuity of the heat flux through the char front: \(\Delta h_p\) is the specific heat of pyrolysis due to the loss of energy by the particle associated with biomass-char conversion and the volatiles released to the gas. \(\Delta h_p\) is defined per unit of biomass mass converted, so \(\dot{m}_h\) in (3.25) is given by:

\[
\dot{m}_h = -4\pi r_c^2 \rho_B \frac{dr_c}{dt} = -4\pi r_c^2 \rho_B v_c \tag{3.26}
\]

in which \(v_c = dr_c/dt\) is the velocity of the char front. From (3.18), (3.19) and the boundary conditions we obtain:

\[
\psi_0 = \phi_0 = T_p \]

\[
2k_C \psi_2 (R - r_c) + k_C \psi_1 = \dot{q} \]

\[-2\phi_2 r_c - \phi_1 = 0 \]

\[-k_B \phi_1 = k_C \psi_1 - v_c \Delta h_p \rho_B. \]

From these equations we can express three coefficients in terms of the fourth, for which we choose \(\phi_1\):

\[
\phi_2 = -\frac{\phi_1}{2r_c} \]

\[
\psi_1 = -\frac{k_B}{k_C} \phi_1 + \frac{v_c \Delta h_p \rho_B}{k_C} \]

\[
\psi_2 = \frac{\dot{q}}{2k_C (R - r_c)} + \frac{k_B \phi_1}{2k_C (R - r_c)} - \frac{v_c \Delta h_p \rho_B}{2k_C (R - r_c)}. \]

Therefore, the virgin biomass and char temperature profiles are given by:

\[
T_C = T_p + \rho_B v_c \Delta h_p - k_B \phi_1 (r - r_c) + \frac{\dot{q} + k_B \phi_1 - v_c \Delta h_p \rho_B}{2k_C (R - r_c)} (r - r_c)^2 \tag{3.27}
\]

\[
T_B = T_p + \phi_1 (r_c - r) - \frac{\phi_1}{2r_c} (r - r_c)^2. \tag{3.28}
\]
Integration of $r^2T$ from $r_c$ to $R$ using (3.20) yields:

$$\frac{d}{dt} \left\{ \int_{r_c}^{R} r^2T \, dr \right\} = -r_c^2T_p \frac{dr_c}{dt} + \int_{r_c}^{R} \alpha_C \frac{\partial}{\partial r} \left( r_2 \frac{\partial T}{\partial r} \right) \, dr$$

$$= -r_c^2T_p \frac{dr_c}{dt} + \alpha_C R^2 \frac{\partial T}{\partial r} \bigg|_{r=R} - \alpha_C r_c^2 \frac{\partial T}{\partial r} \bigg|_{r=r_c}$$

$$= -r_c^2T_p \frac{dr_c}{dt} + \frac{\alpha_C R^2}{k_C} \dot{q} - \frac{\alpha_C r_c^2}{k_C} (v_c \Delta h_p \rho_B - k_B \phi_1) \quad (3.29)$$

in which boundary conditions (3.22) and (3.25) are used. On the other hand, using (3.27) the integral yields:

$$\frac{d}{dt} \left\{ \int_{r_c}^{R} r^2T \, dr \right\} = -r_c^2T_p \frac{dr_c}{dt} + \frac{1}{60k_C} \frac{d}{dt} f(r_c, \dot{q}, v_c, \phi_1) \quad (3.30)$$

where $f(r_c, \dot{q}, v_c, \phi_1) = 9R^4a + 6R^4 \dot{q} - 11R^3ar_c - 9R^3qr_c + R^2 \dot{q}r_c^2 - R^2 ar_c^2 + R \dot{q}r_c^3 - R ar_c^3 + 4ar_c^4 + \dot{q}r_c^4$ and $a = v_c \rho_B \Delta h_p - k_B \phi_1$. Combining (3.29) and (3.30) we obtain from energy conservation in the char region the following equation:

$$\frac{d}{dt} f(r_c, \dot{q}, v_c, \phi_1) = 60\alpha_C \{ R^2 \dot{q} + k_B r_c^2 \phi_1 - r_c^2 v_c \Delta h_p \rho_B \} \quad (3.31)$$

which is the first of the three governing equations in the pyrolysis phase. For $\omega \neq 0$ the volatiles term in (3.21) leads to an additional term in the right-hand side of (3.31):

$$\frac{d}{dt} f(r_c, \dot{q}, v_c, \phi_1) = 60\alpha_C \{ R^2 \dot{q} + k_B r_c^2 \phi_1 - r_c^2 v_c \Delta h_p \rho_B + (1 - \omega) \rho_B v_c C_{vol} r_c^2 (T_s - T_p) \}. \quad (3.32)$$

The second governing equation is obtained by integrating $r^2T$ in the virgin biomass region using (3.27):

$$\frac{d}{dt} \left\{ \int_{0}^{r_c} r^2T \, dr \right\} = r_c^2T_p \frac{dr_c}{dt} + \int_{0}^{r_c} \alpha_B \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) \, dr$$

$$= r_c^2T_p \frac{dr_c}{dt} + \alpha_B r_c^2 \frac{\partial T}{\partial r} \bigg|_{r=r_c}$$

$$= r_c^2T_p \frac{dr_c}{dt} - \alpha_B r_c^2 \phi_1 \quad (3.33)$$

where the derivative of (3.28) is used. On the other hand, the integral of $r^2T$ using (3.28) gives:

$$\frac{d}{dt} \left\{ \int_{0}^{r_c} r^2T \, dr \right\} = r_c^2T_p \frac{dr_c}{dt} + \frac{d}{dt} \left( \frac{1}{15} \phi_1 r_c^4 \right). \quad (3.34)$$

Combining (3.33) and (3.34) we obtain from the energy conservation in the virgin biomass region the second governing equation in the pyrolysis phase:

$$\frac{d}{dt} \left( \frac{1}{15} \phi_1 r_c^4 \right) = -\alpha_B r_c^2 \phi_1. \quad (3.35)$$
The third governing equation in this phase is:
\[
\frac{d}{dt} r_c = v_c \tag{3.36}
\]
which is just the definition of the char front velocity.

The unknowns in the three governing equations are \(T_s, v_c, \phi_1\). The governing equations can be written in matrix form as:
\[
M \cdot \frac{d}{dt} \lambda = \xi \tag{3.37}
\]
with:
\[
\lambda = [T_s, v_c, \phi_1]^T, \quad \xi = \begin{bmatrix} \frac{\partial r_c}{\partial T_s} + \frac{\partial r_c}{\partial \dot{q}} \frac{\partial \dot{q}}{\partial T_s} \phi_1 + 4\phi_1 r_c^3 \frac{\partial r_c}{\partial \phi_1} \phi_1 + 4\phi_1 r_c^3 \frac{\partial r_c}{\partial v_c} \phi_1 \\ 60\alpha_B r_c^2 \phi_1 \\ 60\alpha_C \{R^2 \dot{q} + k_B r_c^2 \phi_1 - r_c^2 v_c \Delta h_p \rho_B + (1 - \omega) \rho_b v_c C_{\text{vol}} r_c^2 (T_s - T_p)\} \end{bmatrix} \tag{3.38}
\]
\[
M = \begin{bmatrix} \frac{\partial r_c}{\partial T_s} + \frac{\partial r_c}{\partial \dot{q}} \frac{\partial \dot{q}}{\partial T_s} & r_c^4 + 4\phi_1 r_c^3 \frac{\partial r_c}{\partial \phi_1} & 4\phi_1 r_c^3 \frac{\partial r_c}{\partial v_c} \\ \frac{\partial \dot{f}}{\partial r_c} (\frac{\partial r_c}{\partial T_s} + \frac{\partial r_c}{\partial \dot{q}} \frac{\partial \dot{q}}{\partial T_s}) + \frac{\partial \dot{f}}{\partial \dot{q}} \frac{\partial \dot{q}}{\partial T_s} & \frac{\partial \dot{f}}{\partial \dot{q}_s} & \frac{\partial \dot{f}}{\partial \dot{q}_c} \\ \frac{\partial \dot{f}}{\partial \dot{q}_s} & \frac{\partial \dot{f}}{\partial \dot{q}_c} & \frac{\partial \dot{f}}{\partial \dot{q}_v} \end{bmatrix} \tag{3.39}
\]

and:

The temperature profile inside a thermally thick particle consists of three regions (see Figure 3.1 (e)). From the core to \(r_t\) the temperature equals \(T_0\), while from \(r_t\) to \(r_c\) and from \(r_c\) to the surface, the temperature is assumed to have a quadratic dependence on \(r\):
\[
\begin{align*}
T(r) &= T_0 \quad \text{for } 0 \leq r \leq r_t \tag{3.41} \\
T(r) &= \phi_2 (r - r_c)^2 + \phi_1 (r - r_c) + \phi_0 \quad \text{for } r_t \leq r \leq r_c \tag{3.42} \\
T(r) &= \psi_2 (r - r_c)^2 + \psi_1 (r - r_c) + \psi_0 \quad \text{for } r_c \leq r \leq R, \tag{3.43}
\end{align*}
\]
whose coefficients have to be determined using the boundary conditions. In the following we show how to derive three governing equations for this phase, without considering char-volatiles heat exchange.

### 3.2.4 Post-pyrolysis heating

This phase is similar to the pre-pyrolysis phase with biomass properties replaced by char properties. Hence, the governing equation is (3.17) with \(\alpha_B\) and \(k_B\) replaced by \(\alpha_C\) and \(k_C\).

### 3.2.5 Thermally thick particle

The temperature profile inside a thermally thick particle consists of three regions (see Figure 3.1 (e)). From the core to \(r_t\) the temperature equals \(T_0\), while from \(r_t\) to \(r_c\) and from \(r_c\) to the surface, the temperature is assumed to have a quadratic dependence on \(r\):
Combining (3.45) and (3.46), we obtain the first governing equation in this phase:

$$T_B(r) = T_p - 2(T_p - T_0) \left( \frac{r_c - r}{r_c - r_t} \right) + (T_p - T_0) \left( \frac{r_{c} - r}{r_{c} - r_{t}} \right)^{2}. \quad (3.44)$$

Using (3.20), the following integral yields:

$$\frac{d}{dt} \left\{ \int_{r_t}^{r_c} r^{2}T_B(r)dr \right\} = r^{2}T_{p} \frac{dr_{c}}{dt} - r^{2}T_{0} \frac{dr_{t}}{dt} + \int_{r_t}^{r_c} \alpha_B \frac{\partial}{\partial r} \left( r^{2} \frac{\partial T_B}{\partial r} \right)dr$$

$$= r^{2}T_{p} \frac{dr_{c}}{dt} - r^{2}T_{0} \frac{dr_{t}}{dt} + 2\alpha_B(T_p - T_0) \frac{r_{c}^{2}}{r_{c} - r_{t}} \quad (3.45)$$

in which (3.44) and (3.3) have been substituted. On the other hand, integration using (3.44) gives:

$$\frac{d}{dt} \left\{ \int_{r_t}^{r_c} r^{2}T_B(r)dr \right\} = \left( \frac{2}{5}T_{0} + \frac{3}{5}T_{p} \right) r_{c}^{2} \frac{dr_{c}}{dt} - \frac{1}{10}(T_{p} - T_{0}) (2r_{c}r_{t} \frac{dr_{c}}{dt} + r_{t}^{2} \frac{dr_{t}}{dt})$$

$$- \frac{1}{15}(T_{p} - T_{0}) (r_{t}^{2} \frac{dr_{t}}{dt} + 2r_{c}r_{t} \frac{dr_{c}}{dt}) - \left( \frac{9}{10} + \frac{1}{10}T_{p} \right) r_{t}^{2} \frac{dr_{t}}{dt} \quad (3.46)$$

Combining (3.45) and (3.46), we obtain the first governing equation in this phase:

$$\frac{d}{dt} h(r_t, r_c) = -60\alpha_B \frac{r_c^2}{r_c - r_t}, \quad (3.47)$$

with $h(r_t, r_c) = 4r_c^3 + 3r_c^2r_t + 2r_cr_t^2 + r_t^3$.

Applying boundary conditions (3.22), (3.24), and (3.25) to (3.43) we obtain the coefficients of the temperature profile in the char region:

$$\psi_0 = T_p \quad (3.48)$$

$$\psi_1 = 2\frac{k_B (r_p - T_0)}{k_C} \frac{T_p - T_0}{r_c - r_t} + \rho_B v_c \Delta h_p \quad (3.49)$$

$$\psi_2 = \frac{\tilde{q}}{2k_C (R - r_c)} - \frac{k_B}{k_C} \frac{T_p - T_0}{(R - r_c)(r_c - r_t)} - \frac{\rho_B v_c \Delta h_p}{2k_C (R - r_c)}. \quad (3.50)$$

In the same way as in the virgin biomass region, the equality of the integrals of $r^{2}T_C(r)$ between $r_c$ and $R$ leads to the second governing equation:

$$\frac{d}{dt} g(r_c, v_c, r_t) = 60\alpha_C \left\{ R^{2} \tilde{q} - r_c^{2} \left( \rho_B v_c \Delta h_p + 2k_B \frac{T_p - T_0}{R_c - r_t} \right) \right\} \quad (3.51)$$

with $g(r_c, v_c, r_t) = \tilde{q}(6R^4 - 9R^3r_c + R^2r_c^2 + R^2r_t^2 + r_t^4) + (2k_B(T_p - T_0)/(r_c - r_t) + \rho_B v_c \Delta h_p)(9R^4 - 11R^3r_c - R^2r_c^2 - R^2r_t^2 - r_t^4)$. The heat exchange between volatiles and char, modeled in equation (3.21), leads to an extra term $(1 - \omega)\rho_B v_c C_{vol} r_c^2(T_s - T_p)$ on the right-hand side of (3.51), representing the heat exchange between volatiles and char. The third governing equation is (3.36), as for a thermally thin particle.

Similarly to the thin particle model, the governing equations can be written in a matrix form and integrated in terms of the dependent variables $\lambda = [T_s, v_c, \phi_1]^T$. 
3.3 Stiffness problem of the equations in the pyrolysis phase

In a DNS with two-way coupling of mass, momentum and energy, it is essential that the properties of both phases (carrier and dispersed) are computed at the same time level. However, the system of equations for the particles in the pyrolysis phase cannot be solved with the same integration method as the one used for the Navier-Stokes equations of the gas. In the early and late phases of pyrolysis, the particle equations are stiff, for both the thermally thick and the thermally thin particle models. This stiffness is caused by the small size of the char region in the early phase and the small size of the virgin biomass region in the late phase. The stiffness of the equations was analyzed using solvers for stiff problems available in MATLAB. The analysis shows that in the early pyrolysis phase the temperature profile in the thin char region is approximately linear (Figure 3.3). Therefore, in order to avoid the stiffness, we assume a linear temperature in the char region if $r_c(t) > \theta R$, with $\theta = 0.9$. In the last section of this chapter we show that $\theta = 0.9$ corresponds to an error smaller than 2%. Instead of solving a system of three equations for the three unknowns $T_s, v_c$ and $\phi_1$, we only solve (3.35) and (3.36) for $T_s$ and $\phi_1$ and find $v_c$ from the condition that $\phi_2 = 0$. Similarly, in the later phase of pyrolysis the temperature in the virgin biomass region is in good approximation constant (Figure 3.4). Coefficient $\phi_1$ is plotted versus time in Figure 3.5. In the late phase of pyrolysis $\phi_1$ goes to zero. Therefore, in this phase it is a good approximation to assume a constant temperature $T_p$ in the virgin biomass region.

These assumptions transform the stiff or basic problem into a non-stiff problem. In Figure 3.6, a comparison of the surface temperature $T_s$ predicted by the non-stiff model and the basic model shows that the assumptions made are accurate. Accuracy depends also on the criteria to switch from a pyrolysis phase to the next one, in particular on the value of $\theta$. We will discuss this in section 3.5. The non-stiff problem is solved using a low storage explicit Runge-Kutta method, whereas the basic problem is solved using the ode15s solver available in MATLAB, designed to solve stiff problems. By applying the approximations in the early and late phases of pyrolysis, the pyrolysis phase is divided into three sub-phases: early pyrolysis stiff sub-phase, regular pyrolysis sub-phase, and late pyrolysis stiff sub-phase. The equations of the second sub-phase are the same as presented before, for the thin and thick particle models. The equations in the other two sub-phases are presented in the following sub-sections.

3.3.1 Early stiff phase of pyrolysis

In this phase the biomass is divided in two regions, the virgin biomass region and the char region. The thermal front has already reached the center of the particle and the char front has only just entered the particle, e.g., $r_c > \theta R$. A typical value of $\theta$ is 0.9, but special attention must be paid to very small particles. The region with linear temperature profile is $r_c < r < R$, where $\psi_2 = 0$. The differences between the non-stiff and the basic model can be reduced using a value of $\theta$ close to 1. However, the choice
for the size of this region depends on the time step used for the time integration. If the time step is too large for a given size of this region, the number of time steps in this phase could become too small, resulting in a large error or even instability (section 3.5). Using the boundary conditions (3.22), (3.23), (3.24), (3.25), and the fact that \( \psi_2 = 0 \) in \( r_c < r < R \) we obtain:

\[
\begin{align*}
 v_c &= \frac{\dot{q} + k_B \phi_1}{\Delta h_p \rho_B} \\
r_c &= f(T_s) = R - \frac{k_C(T_s - T_p)}{\dot{q}}.
\end{align*}
\] (3.52)
Repeating the same procedure of integrating $R^2T$, as shown in Section 3.2.3, we can write:

$$\frac{d}{dt} \left( \phi_1 f^4(T_s) \right) = -15\alpha_B f^2(T_s) \phi_1$$

$$= f^4(T_s) \frac{d\phi_1}{dt} + 4\phi_1 f^3(T_s) \frac{df(T_s)}{dt}$$

$$= f^4(T_s) \frac{d\phi_1}{dt} + 4\phi_1 f^3(T_s) \frac{\dot{q} + k_B \phi_1}{\Delta h_p \rho_B}$$

where we used

$$\frac{df(T_s)}{dt} = \frac{df}{dT_s} \frac{dT_s}{dt}$$

so that the governing equations of this sub-phase (for $r_c > \theta R$) in terms of the unknown $T_s$ and $\phi_1$ are:

$$\frac{dT_s}{dt} = \frac{v_c}{f(T_s)} \frac{df}{dT_s}$$

$$\frac{d\phi_1}{dt} = -\frac{15\alpha_B \phi_1}{f^2(T_s)} - \frac{4\phi_1}{f(T_s)} \frac{\dot{q} + k_B \phi_1}{\Delta h_p \rho_B}.$$

If the particle is thick enough, the pyrolysis will start on the surface before $r_t$ reaches the center. In that case, for $r_c > \theta R$, we assume a linear temperature profile in the char region which leads to the set of governing equations presented in the following sub-section.

### 3.3.2 Thick particle: early stiff phase

In this phase three regions can be identified inside the particle: the char region where the temperature profile is assumed to be linear, the virgin biomass region which is divided by the thermal front $r_t$ in a region with the initial constant temperature and a heating-up region with an assumed parabolic temperature profile. Similarly as in the previous sub-section, from the boundary conditions we obtain:

$$v_c = \frac{\dot{q} - \frac{2k_B (T_s - T_0)}{r_c - r_t}}{\Delta h_p \rho_B}$$

and $r_c$ follows as (3.52). The equations for the unknown $T_s$ and $r_t$ in this phase are:

$$\frac{dT_s}{dt} = \frac{v_c}{f(T_s)} \frac{df}{dT_s}$$

$$\frac{dr_t}{dt} = -\frac{15\alpha_B r_c^2}{r_c - r_t} - \frac{(12r_c^2 + 6r_c r_t + 2r_t^2)v_c}{3r_c^2 + 4r_c r_t + 3r_t^2}.$$

These equations are solved for $r_c > \theta R$ and $r_t > 0$. For very big particles it may happen that $r_c < \theta R$, i.e. the temperature profile in the char region can no longer be approximated by a linear function, while the thermal front has not yet reached the center of the particle, i.e. $r_t > 0$. This case is discussed in the following subsection.
3.3.3 Thick particle pyrolysis with moving thermal front

For very big particles three layers may co-exist as in the previous sub-section, but the linear temperature assumption in the char region no longer holds \((r_c < \theta R)\). In such a case, the problem is described by the equations presented in sub-section 3.2.5.

3.3.4 Late stiff phase of pyrolysis

This phase is the same for the thin and the thick particle model. In this phase we observed that the temperature profile in the virgin biomass region is in good approximation constant and equal to \(T_p\). Therefore, the governing equations are (3.32) and (3.36) with \(\phi_1 = 0\).

A summary of the criteria to switch between the different phases is presented in the next section. The accuracy related to the choice of a value for the criteria is discussed in section 3.5.

3.4 Biomass particle routes undergoing pyrolysis

Previously, we introduced the equations for each phase of a biomass particle undergoing pyrolysis. In Section 3.3 we discussed the stiffness of the problem, which is tackled using approximations for the temperature profiles. With these approximations, we divide the pyrolysis phase in three sub-phases and therefore it is possible to distinguish two possible routes of a thermally thick particle undergoing pyrolysis, which, together with the route of a thermally thin particle, form a total of three possible routes for a pyrolyzing biomass particle.

In order to have a better picture of the various phases, the routes of a biomass particle undergoing pyrolysis are schematically shown in Figure 3.7. The scheme also shows the criteria used to switch between the phases in the numerical code. By \(\epsilon_R\), a small value close to zero is indicated. Initially, the virgin biomass is heated. Afterwards, depending on the particle size and the heat flux applied, the particle may follow three different routes:

1. A thin particle will move to the thin particle pre-pyrolysis phase when \(r_t < \epsilon_R\).
   Secondly, when \(T_s > T_p\), to the early stiff phase of pyrolysis and next, to the pyrolysis phase if \(r_c \leq \theta R\). Then, if \(\phi_1 < \epsilon_{\phi_1}\), the equations of the late stiff phase of pyrolysis are solved. And finally, the particle moves to the post-pyrolysis or char heating phase.

2. A thick particle will move to the thick particle early stiff phase when \(T_s > T_p\).
   Then, if \(r_t \leq \epsilon_R\) before \(r_c \leq \theta R\), the thick particle moves to the early stiff phase of pyrolysis and follows the same route as a thermally thin particle.

3. Otherwise, the thermally thick particle moves to the thick particle pyrolysis with moving thermal front and when \(r_t \leq \epsilon_R\) to the pyrolysis phase. Next, it follows the same route as a thin particle.

The simplified pyrolysis model of Haseli is successfully adapted to be implemented in DNS with a point-particle approach. All necessary steps to make the model suitable for implementation in a DNS code were described. In the next chapter its application in DNS is presented.
3.4 Biomass particle routes undergoing pyrolysis

Figure 3.7. Scheme of the routes of a biomass particle undergoing pyrolysis - in the flow diagram the criterion which occurs first dictates the route followed by the particle.
3.5 Analysis of Pyrolysis Model

In this section a more detailed analysis of the biomass particle pyrolysis model is presented. The aim of this analysis is the development of a more efficient numerical method to solve the system of ordinary differential equations for the biomass particles, which can be coupled to the time integration method applied to the surrounding gas. The efficiency of the numerical method previously described in this chapter is hampered by the stiffness of the system of equations in the beginning and end of the pyrolysis phase and by the singularity of the ordinary differential equation in the beginning of the heating of the virgin biomass.

In the following of this section we discuss only the problems related to the initial phase of the heating of virgin biomass and the initial phase of pyrolysis. The way to deal with the stiffness in the late phase of pyrolysis is treated in sub-section 3.3.4.

3.5.1 Heating of virgin biomass phase

The governing equation (3.9) in the first phase, the heating of a virgin biomass, in which the thermal front moves from the particle surface to the particle center, is written as:

$$\frac{d}{dt}(\dot{q}f(r_t)) = 60\alpha_B\dot{q},$$

where

$$f(r_t) = 6R^2 - 9Rr_t + r_t^2 + \frac{r_t^3}{R} + \frac{r_t^4}{R^2},$$

the position of the thermal front is related to the surface temperature, $T_s$, by:

$$r_t = R - \frac{2k_B(T_s - T_0)}{\dot{q}}.$$  

The heat flux to the particle is given by:

$$\dot{q} = h(T_g - T_s) + \sigma\varepsilon(T_w^4 - T_s^4).$$

All other parameters are constants and described earlier in this chapter. This problem has to be solved with initial condition $T_s(0) = T_0$.

As at the initial time $r_t = R$, it follows that $f(r_t) = 0$ at the initial time, which causes the singularity of this phase of the process. In order to analyze this singularity and find an approximate solution for small times, we make an expansion of the equation for small values of time. To that end we write $T_s(t) = T_0 + \tau(t)$. Then the left-hand side of equation (3.53) can be written as

$$\frac{d}{dt}(\dot{q}f(r_t)) = 40\frac{k_B^2}{\dot{q}_0} \left( \tau^2 - \frac{k_B + R\beta}{R\dot{q}_0} \tau^3 + \mathcal{O}(\tau^4) \right),$$

where we have written $\dot{q} = \dot{q}_0 + \beta\tau + \mathcal{O}(\tau^2)$ with $\beta = -(h + 4\sigma\varepsilon T_0^3)$. The right-hand side of the equation can be written in the form of a Taylor series in $\tau$ as: $60\alpha_B(\dot{q}_0 + \beta\tau + \mathcal{O}(\tau^2))$. 
3.5 Analysis of Pyrolysis Model

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle radius $R$</td>
<td>$1 \times 10^{-4} \text{m}$</td>
</tr>
<tr>
<td>Virgin biomass mass density</td>
<td>$650 \text{ (Kg/m}^3\text{)}$</td>
</tr>
<tr>
<td>Biomass char mass density</td>
<td>$80 \text{ (Kg/m}^3\text{)}$</td>
</tr>
<tr>
<td>Heat transfer coefficient $h$</td>
<td>$152.8 \text{ W/m}^2\text{K}$</td>
</tr>
<tr>
<td>Gas reactor temperature $T_g$</td>
<td>$1400 \text{ K}$</td>
</tr>
<tr>
<td>Wall reactor temperature $T_w$</td>
<td>$1400 \text{ K}$</td>
</tr>
<tr>
<td>Initial particle temperature $T_0$</td>
<td>$300 \text{ K}$</td>
</tr>
<tr>
<td>$\Delta h_p$</td>
<td>$-270 \text{ kJ/kg}$</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>$5.67 \times 10^{-8} \text{ W/(m}^2\text{K}^4)$</td>
</tr>
<tr>
<td>$\epsilon$</td>
<td>$0.9$</td>
</tr>
</tbody>
</table>

Table 3.1. Values used in the simulations.

The lowest order terms in $\tau$ can easily be solved and yield:

$$T_s(t) = T_0 + \sqrt{\frac{3\alpha_B \dot{q}_0^2}{2k_B}} t.$$  

To find the next term in the expansion we write:

$$\tau = b_0 \sqrt{t} + b_1 t^p,$$

where $b_0 = \sqrt{3\alpha_B \dot{q}_0^2/(2k_B)}$ and $p$ and $b_1$ have to be determined. A non-trivial solution is found for $p = 1$, which yields

$$b_1 = \frac{1}{2} \frac{k_B + R\beta}{R \dot{q}_0} b_0^2 + \frac{1}{4} \frac{\alpha_B \beta \dot{q}_0}{k_B^2}.$$  

Figure 3.8 shows the solution for the surface temperature as a function of time during the heating of the virgin biomass phase along with the first and second order approximations in time. Figure 3.9 shows the corresponding solution for the position of the thermal front. The results are obtained for the values indicated in Table 3.1. Similar results are obtained for other values of the parameters in the regime of thermally thin particles. In can be seen that the results of the approximate solutions agree well with the full solution for small values of time and that the second-order terms in the expansion improve the agreement.
Figure 3.8. Surface temperature as a function of time - solution of the complete set of equations (---), first-order solution in time (----), second-order solution in time (-----).

Figure 3.9. Thermal front position $r_t$ as a function of time - solution of the complete set of equations (---), first-order solution in time (----), second-order solution in time (-----).
3.5.2 Pyrolysis phase: first part

The governing equations in the pyrolysis phase are given by:

\[
\frac{df}{dt}(T_s, \phi_1, v_c) = 60\alpha_c \{R^2 q + k_B r_c^2 \phi_1 - r_c^2 v_c \Delta h_p \rho_0 + (1 - \omega) \rho_B v_c C_{vol} r_c^2 (T_s - T_p)\},
\]

(3.54)

\[
\frac{d}{dt}(\phi_1 r_c^4) = -15\alpha_B r_c^2 \phi_1,
\]

(3.55)

and

\[
\frac{dr_c}{dt} = v_c.
\]

(3.56)

In these equations the surface temperature, \(T_s\), the coefficient of the linear term of the quadratic polynomial of \(T(r)\) in the biomass region, \(\phi_1\), and the velocity of the char front, \(v_c\), are used as dependent variables. The position of the char front, \(r_c\), and the heat flux from the surroundings, \(\dot{q}\), can be expressed as functions of the dependent variables according to:

\[
r_c = R - \frac{2k_c (T_s - T_p)}{\rho_0 \Delta h_p v_c - k_B \phi_1 + \dot{q}}
\]

and

\[
\dot{q} = h(T_g - T_s) + \sigma \epsilon (T_w^4 - T_s^4).
\]

Finally:

\[
f(T_s, \phi_1, v_c) = R^4(9a + 6\dot{q}) - R^3 \dot{r}_c(11a + 9\dot{q}) + R r_c^2(\dot{q} - a) (R + r_c) + r_c^4 (4a + \dot{q}),
\]

where \(a = v_c \rho_B \Delta h_p - k_B \phi_1\). All other parameters are constants and described earlier in this chapter.

This set of equations has to be solved with initial conditions:

\[
T_s(0) = T_p,
\]

\[
\phi_1(0) = -\frac{\dot{q}_0}{k_B},
\]

and

\[
v_c(0) = 0,
\]

where:

\[
\dot{q}_0 = h(T_g - T_p) + \sigma \epsilon (T_w^4 - T_p^4)
\]

and we take the initial time of the pyrolysis phase for simplicity, but without loss of generality, equal to 0.

We analyze this system of equations for small values of time. To that end, we write \(T_s(t) = T_p + \tau(t)\) and \(\phi_1(t) = -\dot{q}_0/k_B + \psi(t)\). Moreover, by assuming that the solution of the system of equations is a sufficiently smooth function of \(t\), we can write:

\[
\dot{q} = \dot{q}_0 - (h + 4\sigma \epsilon T_p^3) \tau + \mathcal{O}(\tau^2) \approx \dot{q}_0 + \beta \tau,
\]

where
where $\beta = -(h + 4\sigma T_p^3)$ is a constant. This makes it possible to calculate multidimensional Taylor expansions of the left- and right-hand sides of (3.54-3.56) for small values of $\tau$, $\psi$ and $v_c$. Equation (3.56) is in lowest order of the small parameters equal to:

$$\frac{d\tau}{dt} - \frac{1}{\dot{q}_0} \frac{d}{dt} \left( \beta \tau^2 - \frac{1}{2} k_B \psi \tau + \frac{1}{2} \rho_B \Delta h_p v_c \tau \right) = -\frac{\dot{q}_0}{k_C} v_c. \quad (3.57)$$

This equation shows that $\tau$ is of smaller order of magnitude than $v_c$: If $v_c \sim t^n$, then $\tau \sim t^{n+1}$.

With this knowledge, the lowest order terms in the expansion of equation (3.54) are given by:

$$\frac{d\tau^2}{dt} = \frac{2\alpha C \dot{q}_0}{k_C^2} \left( k_B \psi - \rho_B \Delta h_p v_c + 2\frac{k_c}{R} \right). \quad (3.58)$$

The first and second term on the right-hand side are of lowest order and should cancel in order to satisfy the equation. This implies that to lowest order in powers of time:

$$k_B \psi = \rho_B \Delta h_p v_c. \quad (3.59)$$

Finally, expansion of equation (3.55) gives:

$$\frac{d\psi}{dt} + \frac{1}{k_B} \left( 4\frac{k_C}{R} - \beta \right) \frac{d\tau}{dt} = \frac{15\alpha_B \dot{q}_0}{R^2 k_B} - \frac{15\alpha_B}{R^2} \psi. \quad (3.60)$$

This equation shows that to first order $\psi$ grows linearly in time. From (3.59) it follows that also $v_c$ increases linearly in time to first order and this implies that $\tau \sim t^2$.

Equations (3.57-3.60) also allow the determination of the second order terms in $t$. The results are:

$$T_s = T_p - \frac{15\alpha_B \dot{q}_0^2}{2R^2 \rho_B \Delta h_p k_C} t^2,$$

$$\phi_1 = -\frac{\dot{q}_0}{k_B} + \frac{15}{k_B} \frac{\alpha_B \dot{q}_0}{R^2} t - \frac{15\alpha_B \dot{q}_0 \left( 15 k_C \alpha_B \rho_B \Delta h_p - 4 k_C R \dot{q}_0 + \beta \dot{q}_0 R^2 \right)}{2 \rho_B \Delta h_p k_B R^4 k_C} t^2,$$

and

$$v_c = 15 \frac{\alpha_B \dot{q}_0}{R^2 \rho_B \Delta h_p} t - \frac{15\alpha_B \dot{q}_0 \left( 15 k_C \alpha_B \rho_B \Delta h_p - 2 k_C R \dot{q}_0 + \beta \dot{q}_0 R^2 \right)}{2 R^4 \rho_B^2 \Delta h_p^2 k_C} t^2.$$

Figure 3.10 shows the solution for the surface temperature as a function of time during the first parts of the pyrolysis phase along with the first and second order Taylor approximations in time. Figure 3.11 and figure 3.12 show the corresponding solutions for $\phi_1$ and the velocity of the char front. The results are obtained using the values indicated in Table 3.1. Similar results are obtained for other values of the parameters in the regime of thermally thin particles. In can be seen that the results of the approximate solutions agree well with the full solution for small values of time and that the second-order terms in the expansion improve the agreement.
Figure 3.10. Surface temperature as a function of time - solution of the complete set of equations (---), first-order solution in time (---), second-order solution in time (---), solution of the approximate differential equations (-----).

Figure 3.11. Temperature gradient $\phi_1$ as a function of time - solution of the complete set of equations (---), first-order solution in time (---), second-order solution in time (---), solution of the approximate differential equations (-----).
Moreover, the difference between the full solution and the second-order approximation is proportional to \( t^3 \) for small values of \( t \).

More accurate approximate solutions can be obtained if the set of ordinary differential equations obtained after the second-order Taylor expansion is solved. That means that equation (3.57) without the second term on the left-hand side and equation (3.60) are solved and the right-hand side of equation (3.58) is set to zero to obtain a relation between \( v_c \) and \( \psi \) and \( \tau \). This linear set of first-order differential equations for \( \psi(t) \) and \( \tau(t) \) can be solved analytically, but the solutions are too big to be presented here. The solutions are also included as dotted lines in the figures. They indeed show a much better agreement than the polynomial solutions in time presented above for longer times.

The Taylor expansion of the set of governing equations makes clear why the system of equations cannot be solved with an explicit time integration method, but needs a solver for stiff systems of equations. Indeed, in lowest order equation (3.58) is not an ordinary differential equation, but an algebraic equation which relates \( \psi \) to \( v_c \). In the early stiff phase described in sub-section 3.3.1, the differential equation for the surface temperature is replaced by equation (3.59) to solve the stiffness problem. The analysis presented above shows that this indeed solves the stiffness problem. The set of two resulting ordinary differential equations has a similar character as the set of two linear first-order differential equations mentioned above.

The analytical solution of the linear set of first-order differential equations for \( \psi(t) \) and \( \tau(t) \) shows that one of the eigenvalues of the set of equations is negative and the other positive. The negative eigenvalue is largest in absolute value and is inversely proportional to \( R^2 \) for small values of \( R \), while the positive eigenvalue is inverse proportional to \( R \) for small values of \( R \). For small values of \( R \), the negative eigenvalue is approximately equal to \( \lambda_1 = -15\alpha_B/R^2 \) and the positive eigenvalue \( \lambda_2 \simeq \)
3.5 Analysis of Pyrolysis Model

Figure 3.13. Maximum of the absolute eigenvalues of the system of equations in the pyrolysis phase for $R = 1^{-4}m$.

$-2\dot{q}/(R\rho_b\Delta h_p)$. Note that $\Delta h_p < 0$. It follows that for stable numerical solution with an explicit time integration method the time step should satisfy $\Delta t = cR^2/(15\alpha_B)$ for some constant $c < 1$.

Next, we consider the part of the pyrolysis phase in which we solve the full system of equations. This is the system of three ordinary differential equations for the surface temperature $T_s$, the temperature gradient $\phi_1$ and the char front velocity $v_c$. The maximum possible time step in this phase is determined by the maximum of the absolute eigenvalue of the Jacobian matrix of this system of equations. Figure 3.13 shows the dependence of the maximum of the absolute eigenvalue as a function of the position of the char front for the same parameter values as used above and for a particle radius $R = 1 \times 10^{-4}m$. It appears that for $r_c$ close to the radius of the particle the maximum absolute eigenvalue is inversely proportional to $(R - r_c)^2$.

The maximum possible time step in the pyrolysis phase is inversely proportional to the maximum absolute eigenvalue. Therefore, the figure shows that a larger time step can be chosen if the transition from the early stiff part of the pyrolysis phase, where only two differential equations are solved, to the normal part of this phase is postponed. However, a later transition increases the error made in the stiff part. Figure 3.14 shows the relative error in the time when the char front reaches the radial position $0.2R$ as a function of the char front radial coordinate where the transition is made. The figure shows that indeed the error increases when the transition is postponed. The relative error is well below 1% if the transition is made when the char front position $r_c \geq \theta R$, with $\theta = 0.9$.

The maximum absolute eigenvalue of the Jacobian matrix of the system of equations depends on the parameters of the problem. The most important influence is by the radius of the particle. It appears that the maximum possible eigenvalue is inversely proportional to the square of the radius of the particle. Hence, the time step should be chosen proportional to the square of the radius in order to obtain a stable
explicit time-integration method. Combination of the dependence of the maximum absolute eigenvalue on the radius of the particle and the transition radius yields the following criterion for a stable time step:

$$\Delta t \leq 0.4 \frac{(R - r_{tr})^2}{\alpha C},$$

where $r_{tr}$ is the position of the transition from the stiff part to the normal part of the pyrolysis phase. If an accurate solution is required, $R - r_{tr} \ll R$ and this criterion leads to a time step that is significantly smaller than the maximum possible time step in the stiff part of the pyrolysis phase.

### 3.6 Conclusion

In this chapter it was shown how to derive from Haseli’s model of biomass pyrolysis a model which is suitable for implementation in DNS. The implementation is discussed in the next chapter. The steps necessary to avoid the stiffness of the equations were described in detail. The basic model is therefore modified using assumptions for the temperature profile in certain phases of the process. This makes it possible to solve the equations of the non-stiff model with the same numerical integration method as for the Navier-Stokes equation. This integration method is explained in chapter 4.

In the last part of this chapter, the analysis of the stiffness of the system equations shows that loss in efficiency of the non-stiff numerical method can be improved by solving the initial part of the virgin biomass heating phase with an approximate analytical method, and the first part of the pyrolysis phase using approximate differential equations. The analysis shows that there is a relation between the maximum possible time step and the radius of the particle, when an explicit time integration method is applied.
Biomass pyrolysis in DNS of turbulent channel flow

The model for biomass pyrolysis described in the previous chapter is applied to perform DNS of turbulent channel flow with biomass particles undergoing pyrolysis. The pyrolysis model is suitable for point-particle modeling in DNS of laminar or turbulent flow. The flow regime in burners is typically turbulent for enhancing the flow mixing and therefore the combustion efficiency. We test the biomass pyrolysis model in a turbulent channel flow. A real burner geometry was used in previous work by Ghenai and Janajreh [19], who performed a 2D simulation of co-firing biomass with coal. Ghenai and Janajreh [19] did not solve the turbulent fluctuations directly but used the RANS equations adopting the k-ε turbulence model and a stochastic tracking model to predict particle dispersion. In our formulation the particle-gas interactions are modeled in detail, but the combustion model of the gas and the particles is not included as it was in Ghenai and Janajreh [19]. We restrict to the pyrolysis phase, whose prediction is very important for a good prediction of the combustion phase. Implementation of combustion will be a future extension of this research.

In this chapter we introduce the equations of the carrier, which consists of nitrogen. The absence of oxygen ensures that no reaction occurs other than pyrolysis. The computational model in our simulations is composed of the compressible Navier-Stokes equation for momentum transport, coupled to the continuity equation, and the total energy equation. We consider cases without gravity. The dispersed phase is coupled to the carrier phase by means of source terms reflecting the presence of particles. The two-way coupling involves the momentum, the mass and the heat exchange between gas and particles. During pyrolysis, each particle loses maximum 90% of its mass in the form of volatiles released to the surrounding gas. In this formulation, we assume the same properties for the volatiles and the nitrogen. Therefore, the mass lost by the particles is directly added to the gas, which is considered to be a single component gas, consistent with the assumption of low particle concentrations, and hence relatively low amounts of volatiles released compared to the total system.

The particles are treated in a Lagrangian way. They are modeled using the point-particle approach. Usually, this approach implies the particle to have a uniform
temperature, as in chapter 2. However, the biomass pyrolysis point-particle model presented here is based on a parabolic or doubly parabolic temperature profile along the radius of the particle. To the best of our knowledge, this is the first point-particle model which takes into account thermal diffusion inside the particle. In fact, the model allows point-particle modeling using the surface temperature as key parameter in the equations and, at the same time, it describes the thermal diffusion and the conversion inside the particle. However, more key parameters, such as the char front position or velocity, are needed during some phases in order to fully describe the thermal conversion.

An important parameter in the pyrolysis of biomass is the conversion time, i.e., the time biomass particles need to convert fully to char. This is very important for biomass because of the very high content of volatiles released during pyrolysis. The more accurately pyrolysis is modeled, the better the prediction of the concentration of volatiles which influences the combustion process in the presence of oxygen. We focus on the pyrolysis conversion or devolatization and in particular analyze the effect of the two-way coupling on the devolatization. This is the first study in which a detailed description of the particle-gas interaction during pyrolysis is modeled in a three-dimensional framework. The analysis shows that the conversion time depends on the initial volume fraction (number of particles) and on the size of the particles. We also show here the mass loss history of a realistic biomass particle size distribution for comparing the differences between a two-way and a one-way coupling formulation.

In the following of the chapter, the additional terms in the particle model due the time- and space dependency of the gas temperature are presented. Then, the particle equation of motion used to track the particles is described. Section 4.2 of this chapter is devoted to the gas model. The two-way coupling terms in the gas equations are discussed in section 4.3. Next, the numerical method is described and the set up of the numerical simulations. Finally, the effect of the two-way coupling on the pyrolysis of the particles is studied by an analysis of the results of the DNS for various particle sizes, volume fractions, and for a realistic biomass particle size distribution.

4.1 Particle model

The equations of the biomass pyrolysis model presented in Chapter 3 are derived assuming a constant gas temperature. However, in a turbulent flow the temperature of the gas fluctuates in time and, more in general, it shows a time and space dependency $T = T(x,t)$. Moreover, due to two-way coupling, the presence of particles leads to local changes of the gas temperature in time. Consequently, some extra terms appear in the particle equations for taking into account the time dependency of the gas temperature at the location of a particle. This can be easily understood from Eq. (3.16) in the *pre-pyrolysis phase*. In a gas with constant temperature $T_g$, the term on the left-hand side which is differentiated with respect to time, $T_s R - \frac{1}{3} R^2 \dot{q}$, depends on time only through the time dependence of $T_g$. For a non-constant gas temperature $T_g(t)$, the heat flux $\dot{q} = h(T_g(t) - T_s(t)) + \epsilon \sigma (T_w^4 - T_s(t)^4)$ leads to an extra term in the differential equation for $T_s$. In the following, the additional terms due to the time-dependent gas temperature are presented.
4.1.1 Biomass pyrolysis particle model with time-dependent gas temperature

The heat flux acting on the particle surface is the sum of the convective heat flux from the surrounding gas and the radiative heat flux from the walls (see Eq. (3.5)). The wall temperature is assumed constant but the gas temperature may vary in time. In the following, we show the extra terms arising on the right-hand side of the governing equations presented in chapter 3. These are the terms multiplying $\partial \dot{q} / \partial t$, which denotes $\partial \dot{q} / \partial T_y \partial T_y / \partial t$:

1. In the heating of virgin biomass phase, the governing equation (3.12) becomes:

\[
\frac{d}{dt} (T_s - T_0)^2 = \frac{2 (T_s - T_0)}{f (r_t) \frac{\partial \dot{q}}{\partial T}} - f \frac{\partial \dot{q}}{\partial T} \left( \frac{\partial r_t}{\partial T} + \frac{\partial r_t}{\partial T} \right). \tag{4.1}
\]

2. In the pre-pyrolysis phase, the governing equation (3.17) becomes:

\[
\frac{d T_s}{dt} = \frac{3 \alpha_B \dot{q} + R^2 \frac{\partial \dot{q}}{\partial T}}{R - \frac{1}{2} R^2 \frac{\partial \dot{q}}{\partial T}}. \tag{4.2}
\]

3. In the pyrolysis phase, the right-hand side $\xi$ of the governing equations (3.37) becomes:

\[
\begin{bmatrix}
\dot{\xi} - \frac{\partial f}{\partial r_c} \frac{\partial \dot{q}}{\partial r_c}
-15 \alpha_B r_c^2 \dot{q}_1 - 4 \phi_1 v_c \frac{\partial r_c}{\partial T} \\
\dot{\xi} - \frac{\partial f}{\partial r_c} \frac{\partial \dot{q}}{\partial r_c}
-15 \alpha_B r_c^2 \dot{q}_1 - 4 \phi_1 v_c \frac{\partial r_c}{\partial T}
\end{bmatrix} \tag{4.3}
\]

with $\dot{\xi} = 60 \alpha_C \{ R^2 \dot{q} + k_B r_c^2 \dot{q}_1 - r_c^2 v_c \Delta h_p \rho_B + (1 - \omega) \rho_B v_c C_{vol} r_c^2 (T_s - T_p) \}$. 

4. In the post-pyrolysis heating phase, the governing equation is similar to (4.2) with biomass properties replaced by char properties.

5. In case of a thermally thick particle, the following extra terms appear on the right-hand side of the governing equations (3.47), (3.51), and (3.36), respectively:

\[
\begin{bmatrix}
- \frac{\partial \dot{q}}{\partial r_c} \frac{\partial \dot{q}}{\partial B} \\
- \frac{\partial \dot{q}}{\partial r_c} \frac{\partial \dot{q}}{\partial B} \\
- \frac{\partial \dot{q}}{\partial r_c} \frac{\partial \dot{q}}{\partial B} \\
- \frac{\partial \dot{q}}{\partial r_c} \frac{\partial \dot{q}}{\partial B} \\
\end{bmatrix}, \tag{4.4}
\]

with:

\[
r_c = \frac{1}{2} \left( R + r_t - \frac{A + B}{C} + \left( R - r_t - \frac{A}{B} + \frac{C}{B} \right)^2 + 4 \frac{AC}{B^2} \right)^{1/2}
\]

\[
A = 2k_C (T_s - T_p)
\]

\[
B = \rho_B v_c \Delta h_p + \dot{q}
\]

\[
C = 2k_B (T_p - T_0).
\]

The equations presented are used to compute the particle temperature and mass. In the following the equations to compute the particle position are described.
4.1.2 Particle tracking

The particles are tracked in a Lagrangian way. The particle equations of motion are the same as the water droplet equations presented in chapter 2, with the water properties replaced by the biomass properties. The position of the particle is obtained by integration of the trajectory equation in time:

\[
\frac{dx_i(t)}{dt} = v_i
\]

where \(x_i\) and \(v_i\) are the position and the velocity of the particle, respectively. The particle velocity is computed by integration of equation (2.12) in which, in which we assume that the particles are spherical to good approximation.

4.2 Gas model

In this section the governing equations of the gas (carrier phase) are presented. The equations of the gas contain the terms coupling the gas to the discrete phase (particles), which are specified in the next section.

The gas is treated in an Eulerian way. Due to the high temperature variations involved and the associated large variations of mass density, compressibility effects of the gas are important and we consider the compressible equations. The governing equations for compressible flow are based on conservation laws and can be written as:

\[
\int_V \frac{\partial w}{\partial t} \, dV + \int_{\partial V} \hat{n} \cdot f_c \, dS = \int_{\partial V} \hat{n} \cdot f_v \, dS + \int_V L_{2way} \, dV + \int_V F \, dV.
\]

with \(w(x, t) = [\rho u, \rho v, \rho w, e]^T\) the vector of the dependent variables: \(\rho\) is the mass density of nitrogen, \(u, v\) and \(w\) are the Cartesian components of the velocity vector and \(e\) is the total energy density. Here \(t\) denotes time, whereas \(x = [x, y, z]^T\) is the vector of spatial coordinates. \(V\) is an arbitrary volume with boundary \(\partial V\). Equation (4.6) is the Navier-Stokes equation for a compressible flow with the addition of the coupling terms described later. For channel flow, a driving force \(F\) is added to the streamwise momentum equation in such a way that the total mass flow rate in the streamwise direction is exactly constant. The governing equations express that the rate of change of \(w\) in the volume \(V\) is due to the convective fluxes \(f_c = f_c(w)\), which depend on the dependent variables only, and diffusive fluxes \(f_v = f_v(w, \nabla w)\) through the boundaries of \(V\), which depend on the dependent variables and their first order spatial derivatives, and additionally due to the presence of particles. The divergence theorem was used to transform the volume integrals of the diffusive and convective fluxes into surface integrals, with \(\hat{n}\) the unit vector normal to the surface \(\partial V\) pointing in the outward direction. The two-way coupling terms \(L_{2way}\) in the governing equations for the gas phase satisfy the requirement that they only transfer mass, momentum and internal energy from one phase to the other.

All equations in the code are in non-dimensional form, for which a reference temperature, mass density, velocity and length are chosen. This leads to the non-dimensional parameters Reynolds number \((Re = puL/\mu())\), Prandtl number \((Pr = \rho C_p \mu /\kappa)\),
4.2 Gas model

c\_p\mu/k) and Mach number (M = v/v_{\text{sound}}). The convective flux equals:
\[
f_c = \begin{bmatrix} \rho u^2 + p & \rho uv & \rho uw \\ \rho uv & \rho v^2 + p & \rho vw \\ \rho uw & \rho vw & \rho w^2 + p \\ (e + p)u & (e + p)v & (e + p)w \end{bmatrix},
\]
where \( p \) equals the pressure, which is related to the dependent variables by the equation of state:
\[
e = \frac{p}{\gamma - 1} + \frac{1}{2} \rho (u^2 + v^2 + w^2)
\]
and \( \gamma \) is the ratio between the specific heat at constant pressure and the specific heat at constant temperature. For nitrogen \( \gamma = 1.4 \).

For a Newtonian fluid the viscous flux vector is given by:
\[
f_v = \begin{bmatrix} \sigma_{11} & \sigma_{12} & \sigma_{13} \\ \sigma_{21} & \sigma_{22} & \sigma_{23} \\ \sigma_{31} & \sigma_{32} & \sigma_{33} \\ 0 & 0 & 0 \\ \sigma_{k1} u_k - q_1 & \sigma_{k2} u_k - q_2 & \sigma_{k3} u_k - q_3 \end{bmatrix},
\]
Here, \( \sigma_{ij} \) is the viscous stress tensor and \( \mathbf{q} = [q_1 \ q_2 \ q_3]^T \) the heat flux vector. They are given by:
\[
\sigma_{ij} = \frac{\mu(T_g)}{Re} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \frac{\partial u_k}{\partial x_k} \delta_{ij} \right)
\]
and
\[
q_j = -\frac{\mu(T_g)}{(\gamma - 1) \text{RePrM}^2} \frac{\partial T_g}{\partial x_j}.
\]
In these expressions summation over repeated indices is used and \( \delta_{ij} \) is the Kronecker tensor. The gas temperature is denoted by \( T_g \) and related to the dependent variables by the ideal gas law:
\[
T_g = \gamma M^2 \frac{p}{\rho}.
\]
The dynamic viscosity of nitrogen is in good approximation given by Sutherland’s law [51]:
\[
\mu(T_g) = T_g^{3/2} \frac{1 + C}{T_g + C},
\]
with \( C = 120/T_{\text{ref}} \) a constant.
4.3 Two-way coupling terms

The two-way coupling terms in the governing equations of the gas phase do not change the total water mass, total momentum and total energy in the system. They only transfer mass, momentum and energy from one phase to the other. Moreover, we will assume that the two-way coupling terms act as a point force in the governing equations for the gas.

The two-way coupling term in the Navier-Stokes equation for the gas phase (4.6) can be written as:

\[
\int_V \mathbf{L}_{2\text{way}} \, dV = - \sum_{i \in V} \frac{d}{dt} \begin{bmatrix}
  m_i u_i \\
  m_i v_i \\
  m_i w_i \\
  m_i e_i 
\end{bmatrix},
\]

(4.14)

where the sum is taken over all particles within the volume \( V \). The first three components are the momentum exchange between the two phases. The change in momentum is expressed by (2.11). The fourth component is the mass given by the particle to the gas during pyrolysis in the form of volatiles. We assume that the volatile properties are the same as the properties of nitrogen. The mass rate of change or mass loss is:

\[
\frac{dm_i}{dt} = 4\pi (\rho_B - \rho_C) r_c^2 v_c.
\]

(4.15)

The fifth component is the two-way coupling term of the energy, which is the sum of three contributions:

\[
\frac{d}{dt} e_i = h_m A_i (T_g - T_i) + c_{v,f} T_p \frac{dm_i}{dt} - \frac{d}{dt} \left( \frac{1}{2} m_i |v_i|^2 \right).
\]

The first term is the convective heat transfer between gas and particle, which is proportional to the relative temperature, the particle surface area \( A_i \) and the heat transfer coefficient as defined by Bird et al. [7]. The second term is the energy associated to the volatiles given to the gas by the particle during pyrolysis. The volatiles are formed at the char front where the temperature is \( T_p \). The change in the volatiles temperature due to this interaction will be further investigated. In this work, it is assumed that the temperature of the volatiles released is \( T_p \) and the heat capacity of the volatiles and gas is equal. The third term is the change in kinetic energy which is expressed by the drag force and the mass loss of the particle:

\[
\frac{d}{dt} \left( \frac{1}{2} m_i |v_i|^2 \right) = m_i \mathbf{v}_i \cdot \frac{d\mathbf{v}_i}{dt} + \frac{1}{2} |\mathbf{v}_i|^2 \frac{dm_i}{dt}
\]

in which \( d\mathbf{v}_i/dt \) is given by (2.12).

In the next section the numerical method will be described, including the way in which the coupling terms are evaluated.
4.4 Geometry and boundary conditions

The computational domain of the channel flow is a channel with size $4\pi H$ in streamwise direction and $2\pi sH$ in spanwise direction, where $H$ is half the channel height (see figures 4.1). For the streamwise, wall-normal, and spanwise directions the notation $x$, $y$, and $z$ is used, respectively. There are $128^3$ control volumes in total. In both homogeneous directions (spanwise and streamwise) the grid spacing is uniform. In the wall-normal direction, the grid points are clustered near the walls in order to resolve the boundary layer. Periodic boundary conditions are applied in the streamwise and spanwise directions. The no-slip condition is applied on the walls and the wall temperature is kept constant. For the particles, periodic conditions are applied in the homogeneous flow directions, which means that when a particle leaves the domain in one of these directions, it re-enters the domain from the other side with the same properties. If a particle approaches a wall within a distance of its radius, rules of elastic collision of a hard spherical particle are applied without heat transfer.

4.5 Numerical method

We sketch the main elements of the numerical approach applied in the pyrolysis biomass particle model. First, the time integration method for both the continuous and discrete phases is presented. Next, the spatial discretization of the gas phase is explained. Finally, the numerical implementation of the two-way coupling terms is discussed and some aspects of the parallelization used to reduce the simulation time are explained.

4.5.1 Time integration

The time stepping method applied is a low-storage second-order accurate Runge-Kutta method of the form:

$$u^{(j)} = u^{(0)} + \beta_j \Delta t f(u^{(j-1)}),$$  \hspace{1cm} (4.16)
for \( j = 1, 2, 3, 4 \), where \( u^{(0)} \) is the variable at time \( t \) and \( u^{(4)} \) the variable at time \( t + \Delta t \), and \( f \) is evaluated at the previous Runge-Kutta stage. The particle equations are in the form of an ordinary differential equation:

\[
\frac{du}{dt} = f(u) \tag{4.17}
\]

while the gas equation assumes this form after the spatial discretization is applied, with the sum of convective, diffusive and coupling terms denoted by \( f \). For this Runge-Kutta scheme the solution needs to be stored at the previous time and the current stage only. The Runge-Kutta coefficients are: \( \beta_1 = 1/4 \), \( \beta_2 = 1/3 \), \( \beta_3 = 1/2 \) and \( \beta_4 = 1 \).

### 4.5.2 Finite volume method

The spatial discretization is based on a finite volume method. To this end the geometry is divided into small rectangular cells. In case of channel flow, the walls of the channels coincide with the boundaries of the cells and the variables are stored in the centers of the cells. We solve the governing equations (4.6) for each cell, taking \( V \) as the cell volume. If we identify the value of the dependent variables as the averages over a cell the result for cell \((i, j, k)\) is:

\[
\frac{dw_{i,j,k}}{dt} \frac{V_{i,j,k}}{w_{i,j,k}} = \frac{N_{x,i+1/2,j,k} - N_{x,i-1/2,j,k} + N_{y,i,j+1/2,k} - N_{y,i,j-1/2,k} + N_{z,i,j,k+1/2} - N_{z,i,j,k-1/2}}{2} + (L_{\text{2way}})_{i,j,k} + F_{i,j,k}, \tag{4.18}
\]

where \( V_{i,j,k} \) is the volume of the cell and \( N_{x,i+1/2,j,k} \) is the integral of the flux in the \( x \)-direction over the right boundary face and similar for the other fluxes. The two-way coupling term is the sum of the contributions of all particles present in \( V_{i,j,k} \). The matrix form of the gas equations (4.6) helps to identify the fluxes which need to be calculated at the interface of two adjacent control volumes. Since all fluxes except those on the boundaries of the geometry appear in two cells, once with a plus and once with a minus sign, this numerical method is conservative. All fluxes are calculated as the product of the surface area of the boundary face of the cell and an estimate of the value of the flux at the midpoint of this face.

For the convective flux which only involves the dependent variables and no spatial derivatives, first the values of \( w \) in the center of a cell face are found by averaging over the two nearest neighboring cell centers. This means:

\[
w_{i+1/2,j,k} = \frac{1}{2}(w_{i,j,k} + w_{i+1,j,k}).
\]

Next the pressure is determined from these five values from its definition (4.8) and the fluxes are determined according to (4.7). This method is second-order accurate.

The calculation of the viscous fluxes is more complex since it involves calculation of spatial derivatives of velocity components and temperature. First the velocity components and the temperature are calculated in all cell centers. Then all necessary derivatives are calculated in the midpoints of the cell faces by using the smallest stencil possible. For example:

\[
\frac{\partial w}{\partial x} \bigg|_{i+1/2,j,k} = \frac{w_{i+1,j,k} - w_{i,j,k}}{\Delta x}, \tag{4.19}
\]
where $\Delta x$ is the grid spacing in the $x$-direction, or

$$
\left. \frac{\partial w}{\partial x} \right|_{i,j+1/2,k} = \frac{w_{i+1,j,k} - w_{i-1,j,k} + w_{i+1,j+1,k} - w_{i-1,j+1,k}}{4 \Delta x}.
$$

Boundary conditions in the stream- and spanwise direction are periodic. This makes it easy to apply the discretization methods to all cells. In the wall-normal direction the faces of the cells neighboring the walls coincide with the wall. For the convective flux the only non-zero flux is then equal to the pressure. The pressure on the wall is found from (4.8) with velocity equal to zero and the average value of the energy density between the first cell inside the geometry and the dummy cell outside the geometry. The energy density in this dummy cell is set equal to the energy density in the first cell. For calculation of the viscous fluxes in the cell faces coinciding with a wall, the wall-normal derivatives of the three velocity components are needed. They are found from the no-slip boundary condition and the values in the first cell. The wall boundary condition of the energy equation is obtained from the given constant temperature of the walls.

### 4.5.3 Two-way coupling terms

In the finite volume method for the fluid adopted here the two-way coupling term (4.14) is added to the right-hand side of (4.6) accounting for all particles present in the cell. The quantities in (4.14) are already computed in the particle equations, which simplifies the implementation of these terms. The properties of the gas at the particle location need to be computed because they appear in the particle equations and in the two-way coupling terms. To this end a trilinear interpolation is employed, which uses the values of the gas properties in the control volume where the particle is located and in the neighboring control volumes. Once these terms are computed, their contribution is added to the control volume in which the particle is located.

The control volume in which a particle is located can be identified from the coordinates of the center of the particle. The coupling terms stemming from this particle will be included only in the gas equations of this control volume. This implementation is based on the assumption the particle size is much smaller than the Kolmogorov scale of the flow, which is a necessary requirement for treating the particle as a point-particle. However, in the channel flow it may happen that the particle diameter is larger than the height of the grid cell close to the wall, since the grid is non-uniform in the wall-normal direction due to the clustering of grid points in the boundary layer. Also in such a case, we attribute the two-way coupling terms only to the cell where the particle center is, although it might be more accurate to distribute the contribution over more than one cell.

### 4.6 Parallelization

The program is parallelized with MPI. Every processor runs the program and solves the equations on part of the domain. For the fluid the domain is divided into 16 blocks, 4 in the $x$-direction and 4 in the $z$-direction. All blocks have the same size.
Moreover, all blocks have on all sides an extra row of dummy cells, so that all fluxes can be calculated with information of the block. After every stage of the Runge-Kutta scheme the values in the dummy cells are updated. To that end, every block knows the numbers of the neighboring processors.

Particles are distributed uniformly over the 16 processors and particles stay on the same processor. That means that the fluid variables at the particle location have to be calculated on the correct processor. To that end the necessary information is sent from the processor where the particle is to the processor where the fluid information is and back.

For large numbers of particles, the intense data traffic between the processors may slow down the computation, depending on the architecture of the computer. A way to avoid this is to store the particles in the same processor where the gas properties in their grid cells are stored. The implies that after every Runge-Kutta stage the particles that have left their block should be transferred to the processor treating the neighboring block. This method reduces the data traffic, but can lead to small load imbalance.

### 4.7 Set up of the simulations

Simulations are performed at frictional Reynolds number approximately $Re_\tau = 150$, based on friction velocity $u_\tau = \sqrt{\tau_w/\rho_g}$ and half the channel height, where $\tau_w = \mu g \partial \langle u \rangle / \partial y |_{y=\pm H}$ is the wall shear stress and $\langle u \rangle$ the mean streamwise velocity component, averaged over the two homogeneous directions and time. The initial volume fraction varies from simulation to simulation but it is always smaller than $O(10^{-3})$, so that particle-particle collisions can be ignored but two-way coupling might be relevant [13].

In Table 4.1 the properties of the virgin biomass and the char are shown. The virgin biomass density is about eight times larger than the char density. Therefore, a biomass particle loses most of its mass during conversion, releasing large amounts of volatiles. The constants in the radiative heat flux are $\sigma = 5.67 \times 10^{-8}$ W/(m² K⁴) and $\epsilon = 0.9$. The value of the specific heat of pyrolysis is $\Delta h_p = -270$ kJ/kg.

The flow is initialized by a turbulent velocity field in the statistically steady state obtained from a simulation without particles. The initial temperature of the gas is approximately constant at 1400 K, whereas the initial temperature of the particles equals 300 K and is constant within the particle. The wall temperature is kept constant at 1400 K [9]. Initially, particles are randomly and uniformly distributed and the particle velocity is set equal to the gas velocity at the particle location. In the initial transient phase the temperature changes because of the heat flux $\dot{q}$ which

<table>
<thead>
<tr>
<th></th>
<th>Mass density (kg/m³)</th>
<th>Thermal conductivity (W/m°C)</th>
<th>Specific heat capacity (J/kg°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>biomass</td>
<td>650</td>
<td>0.25</td>
<td>2500</td>
</tr>
<tr>
<td>char</td>
<td>80</td>
<td>0.1</td>
<td>1100</td>
</tr>
</tbody>
</table>

Table 4.1. Properties of the virgin biomass and the biomass char used in the simulations.
transfers energy from the gas and from the walls to the colder particles by convection and radiation. Using these settings we simulate an experiment of biomass particles undergoing pyrolysis in a turbulent channel flow with initial isothermal temperature and constant temperature of the walls.

4.8 Results

The conversion time of pyrolysis is the time required by the particles to be converted from biomass to char. It includes the time needed to warm the particles from their initial temperature to the pyrolysis temperature. In the results shown in this chapter, the conversion time is evaluated at 99% conversion of virgin biomass matter. Results evaluated at 95% of conversion are qualitatively the same.

In sub-section 4.8.1 and 4.8.2, we investigate the effect of varying volume fraction and particle size on the conversion time. In sub-section 4.8.3, we compare the results of the simulations with one-way and two-coupling for a realistic biomass particle size distribution (Rosin-Rammler distribution).

4.8.1 Changing volume fraction

We performed simulations with different numbers of particles in order to analyze the dependence of the pyrolysis conversion time on the particle volume fraction. The diameter of all particles was 0.7 mm. The particle Stokes number changes from 40 to 5 during pyrolysis, due to the mass loss. The results in Figure 4.2 show that with two-way coupling the conversion time increases with increasing volume fraction and it is constant with one-way coupling. The two-way particle-gas interaction affects the gas temperature because of the convective heat exchange: more particles extract more heat from the gas compared to simulations with fewer particles. This results in a slower particle heating and increases the pyrolysis conversion time. In the one-way coupling simulations, the temperature is not affected by the presence of particles. In terms of heat exchange, the comparison between one-way and two-way coupling in Figure 4.2 shows that for this case the particle feedback to the gas is important for accurately predicting the conversion time.

The effect of two-way coupling can be observed by studying the time evolution of the temperature profiles averaged over the homogeneous directions. In Figure 4.3 the averaged temperature profiles are shown at different instants of time, as functions of the wall-normal coordinate. The volume fraction $\phi$ is $2.55 \cdot 10^{-4}$. Initially, the gas is much warmer than the particles, so that the particles subtract energy from the gas and from the walls. Therefore, the gas starts to cool down, in the center of the channel more than near the walls because the walls are kept at constant temperature. After a certain time (around $t = 0.32$ s), particles become warmer than the gas because they are continuously heated by the radiative flux from the walls. Therefore, the convective heat exchange changes direction and goes from the particles to the gas. This is due to the lack of a heat source for the gas apart from the heat diffused from the hot walls. In fact, nitrogen does not allow any reaction to take place except for pyrolysis. When the particles are completely converted to char, the char heating continues until the
Figure 4.2. Conversion time at different volume fractions with particle diameter 0.7 mm and initial nitrogen temperature 1400 K - one-way coupling (×), two-way coupling (●).

Figure 4.3. Averaged gas temperature (—) and particle temperature (----) at different instants of time.

particle temperature reaches the wall temperature.
### 4.8.2 Changing particle size

In order to investigate the effect of particle size on the conversion time, we performed simulations with constant volume fraction and varying particle diameter. The volume fraction \(2.55 \times 10^{-4}\) is in the range where particle collisions can be ignored but two-way coupling is relevant \([13]\). Figure 4.4 shows that the conversion time \(t_{\text{conv}}\) increases with increasing particle diameter. The effect of two-way coupling is more clear using a logarithmic scale in which it is visible that \(t_{\text{conv}} \propto R^{1.5}\). The log-log plot shows also that the relative increase of \(t_{\text{conv}}\) is larger for small particles. This means that the two-way coupling effect on the gas is reduced if, at same volume fraction, the particles are bigger. This is explained by the larger total surface area of smaller particles compared to bigger particles with the same volume fraction and by the fact that the heat transfer coefficient is inversely proportional to the diameter of the particle (Eq. refeq hm).

### 4.8.3 Rosin-Rammler size distribution

In practical applications of co-firing of biomass and coal, the probability density function of biomass particle diameter follows a Rosin-Rammler distribution \([48]\). This function is defined as follows:

\[
\frac{k}{\lambda} \left( \frac{d_p}{\lambda} \right)^{k-1} e^{-(d_p/\lambda)^k} \tag{4.21}
\]

with \(d_p\) the particle diameter and \(k\) and \(\lambda\) the shape and scale factors, respectively.

In Figure 4.5 the particle size distribution of beech wood that we use is shown, with \(k = 1.4\), \(\lambda = 0.9\) and the diameter in millimeter. In reality only few particles are larger than 3 mm. This Rosin-Rammler distribution is a good approximation for small particles, but it overpredicts the number of larger particles. Therefore, we
truncated it at 3 mm. Moreover, for particles larger than 3 mm the point-particle assumption becomes less accurate.

The presence of very small particles requires a very small time step because of the stability restriction associated with the explicit time-integration method (section 3.5). Including very small particles would lead to very long computational times. Therefore, we also bounded the size distribution from below. Since it is not known which particle size can be neglected, we performed four simulations in which the cut-off of the particle size distribution was set at 0.4, 0.3, 0.2 and 0.15 mm in order to investigate whether it is possible to disregard very small particles without noticeably influencing the results and saving computational time. The volume fraction of biomass in these simulations is $5.22 \times 10^{-5}$, equivalent to 1000 kg of biomass for 7419 kg of air, corresponding to 20% of air excess with respect to the stoichiometric particles/gas mass ratio (1000/6360).

In the absence of very small particles the pyrolysis and therefore the particle mass loss starts somewhat later, as Figure 4.6 shows. The lower bound for particle diameters implies that almost 12% of the particles present in the distribution is ignored in the simulations, but these ignored particles constitute a negligible mass fraction, less than 0.1% of the total mass of biomass. Hence, it can be concluded that the lower bound will have a negligible influence on the characteristic quantities of the pyrolysis, such as the mass loss history and the conversion time. A cut-off at 0.2 mm or less hardly influences the results. According to [13], the volume fraction used is in the one-way coupling range, where the coupling terms in the momentum equation can be ignored. The results in Figure 4.8.3 demonstrate that the same holds for the coupling terms in the gas temperature equation, as the differences between one-way coupled and two-way coupled simulations are small.

The effect of two-way coupling in the temperature field becomes important at higher volume fractions. Increasing $\phi$ by a factor of 20 ($\phi = 1.044 \times 10^{-3}$) shows that the particle feedback to the gas delays the conversion time with respect to the same simulation with one-way coupling (Figure 4.8.3).
4.8 Results

Figure 4.6. *Biomass mass versus time at different cut-off values* - 0.4 mm (---), 0.3 mm (---), 0.2 mm (----), and 0.15 mm (-----).

Figure 4.7. *Volume fraction $5.22 \times 10^{-5}$* - *Biomass mass loss: one-way (---) and two-way coupling (--).*
Figure 4.8. Volume fraction $\phi = 1.044 \times 10^{-3}$ - Biomass mass loss: one-way (---) and two-way coupling (-----).

4.9 Conclusion

The biomass pyrolysis model is successfully implemented in DNS of turbulent channel flow. The heat and mass exchange influence the biomass pyrolysis conversion and thanks to this formulation their effect can accurately be quantified. The conversion time increases with increasing volume fraction and/or particle size. The particle feedback to the gas is important only at $\phi > 10^{-4}$. In case of a Rosin-Rammler biomass size distribution, the particle feedback to the gas has a minor effect on both the velocity and temperature field of the gas. The reason of that is the low volume fraction of the biomass particles. The very small particles of this distribution have a minor effect on the conversion time, therefore the particles with diameter smaller than 0.2 mm can be ignored.

The model we have presented here is applied to coal particles as well. This application is presented in the next chapter, where the results of simulations with both coal and biomass particles undergoing pyrolysis are presented and discussed.
Coal and biomass co-pyrolysis in DNS of turbulent channel flow

5.1 Introduction

In this chapter the simplified pyrolysis model developed for biomass particles is used to simulate the pyrolysis of coal particles and to combine these two in order to study the so called co-pyrolysis. Similarly to co-firing, in recent studies [59, 26, 52] it has been shown that co-pyrolysis provides an opportunity to improve the utilization of coal and to reduce the CO$_2$ emissions. The production of fuels from coal is limited by the low hydrogen-carbon ratio of coal [59]. The missing hydrogen can be supplied by biomass, which is generally hydrogen-rich. Co-pyrolysis shows a synergetic effect which yields more pyrolysis products, in the form of volatiles, since the coal can be more efficiently converted into volatiles using the hydrogen supplied by the biomass.

The co-pyrolysis synergy depends on the blending ratio and heating rate. The blending ratio is the ratio between the amount of coal which is substituted by biomass and the original amount of coal. The blending ratio can be mass-based, if coal is replaced by the same biomass mass, or energy-based, if the total heating value is kept the same. We investigated the synergetic effect by varying the blending ratio keeping the total heating value constant.

Varying the reactor temperature is theoretically possible, but our present simplified biomass model does not account for the different behavior of the biomass at high and low heating rates. For coal, the amount of volatiles does not significantly increase with increasing heating rate, while for biomass higher heating rates yield more volatiles [43]. In order to include this effect in the model, it would be necessary to establish a correlation between the final mass density of the biomass char and the reactor temperature. Therefore, we restrict our investigations to high heating rates and corresponding high reactor temperatures.

In the next section we discuss the size distributions of coal and torrefied biomass particles, pointing out some issues which need to be solved and the solutions we adopt for these. Next, in section 5.3 we show the results of co-pyrolysis for different blending ratios. Finally, in section 5.4 we discuss the implementation of the co-pyrolysis synergy.
84 Coal and biomass co-pyrolysis in DNS of turbulent channel flow

<table>
<thead>
<tr>
<th></th>
<th>Mass density (kg/m³)</th>
<th>Thermal conductivity (W/m²°C)</th>
<th>Specific heat capacity (J/kg°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>coal</td>
<td>850</td>
<td>0.30</td>
<td>1260</td>
</tr>
<tr>
<td>coal char</td>
<td>600</td>
<td>0.15</td>
<td>1000</td>
</tr>
<tr>
<td>torrefied biomass</td>
<td>650</td>
<td>0.25</td>
<td>2500</td>
</tr>
<tr>
<td>biomass char</td>
<td>190</td>
<td>0.1</td>
<td>1100</td>
</tr>
</tbody>
</table>

Table 5.1. Properties of the coal and the torrefied biomass used in the simulations.

in the model and analyze its effect for various choices of the blending ratio and particle size distribution.

5.2 Coal and biomass particles size distribution

In this section the size distributions of coal and torrefied biomass are presented and the issues related to the implementation of the size distributions in the code are discussed. First, the coal particle distribution is presented, and then the torrefied biomass particle distribution.

5.2.1 Coal particle size distribution

Coal and biomass particle diameter distributions are modeled using a Rosin-Rammler size distribution function, as introduced in section 4.8.3. For coal, we use the cumulative size distribution depicted in figure 5.1, based on data made available by DNV KEMA [9]. Its equivalent probability size distribution function is shown in figure 5.2. The presence of very small particles presents a problem in our simulations since the maximum possible time step to solve the particle equations with the explicit Runge-Kutta method introduced in chapter 4 is proportional to the square of the particle diameter. A small particle requires a small time step which leads to a significantly longer computing time.

In the following we investigate the effect of various values of a cut-off diameter on the mass loss of coal particles during pyrolysis. The simulations were performed using the code described in chapter 4 with the same set up. The biomass particle properties were substituted by the coal properties specified in Table 5.1. The enthalpy of pyrolysis of coal $\Delta h = -418,400$ kJ/kg from Fletcher [17] was used. The coal diameters were generated according to the size distribution of figure 5.1 and the cut-off value of the size distribution was ranging from 30 to 70 µm. For particles smaller than 60 µm the criterion to switch from the early stiff phase to the normal pyrolysis phase was relaxed to $r_c = 0.8 R$. In this a way, the same time step $\Delta t = 3.4189 \times 10^{-5}$ s as in chapter 4 can be used with a relative error at the end of the normal pyrolysis phase smaller than 2% (see figure 3.14).

For a coal particle with diameter 50 µm, a time step $\Delta t = 3.4189 \times 10^{-5}$ s implies that the whole pyrolysis process is finished in approximately 100 time steps. Therefore, we need to verify that the process is described with sufficient accuracy using this time step. Figure 5.3 shows that the surface temperature of a coal particle is almost
the same if a ten times smaller $\Delta t$ is applied. Differences are smaller than 15K, which is about 2%. Therefore, we conclude that $\Delta t = 3.4189 \times 10^{-5}$s yields sufficiently accurate results for the range of particle diameters we consider. For a much smaller particle it may happen that this $\Delta t$ is too large to accurately capture the four phases of the pyrolysis model.

Figure 5.4 shows the mass loss history for different values of the cut-off diameter. The lines almost overlap, indicating that the size distribution is cut in a range where the particles do not considerably influence the pyrolysis time. A close-up view of the mass loss history in the region between the lines of 95% and 99% of the total coal mass conversion shows that there is no systematic trend of the results with the cut-off diameter (figure 5.5). On the other hand, the close-up view of the initial phase in figure 5.6 shows the systematic delay of the mass loss start as a function of the cut-off value: smaller particles pyrolyze earlier than larger particles. As a result, they affect the early phase of pyrolysis but this does not influence the results later in time.

Although figure 5.2 shows that most of the coal particles are not included in the simulation if a cut-off diameter of 50$\mu$m is adopted, the mass loss history displayed in figures 5.4-5.6 shows that the effect of this on the pyrolysis time and mass loss history is very small. This can be explained by the volume distribution function of coal particles shown in figure 5.7, which indicates that the missing particles constitute only 3% of the volume of all coal particles. We conclude that a cut-off diameter of 50$\mu$m, which will be used in this chapter, is a good compromise between computational cost and accuracy of the solution.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{cumulative_distribution.png}
\caption{Cumulative distribution function of coal particle diameter. Rosin-Rammler distribution (---) and experimental data (•) [9].}
\end{figure}
The volume fraction we used in the simulations presented above is $2.88 \times 10^{-5}$, which corresponds to 1 kg of coal for 10.282 kg of air$^1$. This mass fraction is based on 20% air excess with respect to stoichiometric conditions. The number of particles needed to obtain this volume fraction using the particle size distribution in figure 5.2 (with cut-off at 50µm) is approximately 51,000,000 which is prohibitive in terms of computational cost. In order to circumvent this problem, we investigated whether it is possible to solve the particle equations only for a reduced number of particles. In this way every particle represents a number of real particles with the same properties (i.e. location, velocity, temperature, etc.). We name the particles that are solved *effective particles* and the number of particles each of them represents the *particle factor* $\zeta$. In the numerical simulation this method is easily implemented by multiplying the two-way coupling terms in the gas equations with $\zeta$.

Intuitively, it is clear that this method will only be accurate if the number of effective particles does not differ much from the number of computational cells in the numerical method for the gas. Indeed, the two-way coupling terms are equal if two similar particles are present in one cell, or if one effective particle with the same properties and $\zeta = 2$ is present there.

This method is analyzed in figure 5.8 by comparing the mass loss history using different particle factors. The predictions with $\zeta \leq 50$ are well in agreement with each other, while for $\zeta > 50$ the predictions deviate. In figure 5.9, the close-up view in the region between the lines of 95% and 99% of the total coal mass conversion shows that the conversion time predicted with $\zeta = 200$ is better than with $\zeta = 100$. We attribute this to coincidence. Moreover, both lines for $\zeta = 100$ and $\zeta = 200$ significantly differ from the others.

---

$^1$Based on coal mass density $\rho_{cc} = 850$ kg/m$^3$ and air density at 1400 K.
5.2 Coal and biomass particles size distribution

Figure 5.3. Surface temperature of a coal particle with diameter 50µm in uniform temperature of 1400K for different time steps. Δt = 3.4189 × 10⁻⁵ s (—) and Δt = 3.4189 × 10⁻⁶ s (—) [9].

The value of the maximum particle factor depends on the ratio between the number of effective particles and the number of control volumes. Increasing the volume fraction allows a higher value for ζ. Using twice the volume fraction 2 × 2.88 × 10⁻⁵, we compared the mass loss history for particle factors 20, 50, and 100 finding a good agreement, as shown in figure 5.10. However, a too large ζ may lead to instability if a large particle is located close to the wall, where the control volumes are smaller than in the rest of the channel. In that case, a large ζ leads to large two-way coupling terms, resulting in oscillatory gas properties.

5.2.2 Torrefied biomass particle size distribution

Next, we turn our attention to biomass particles and we consider torrefied biomass. The size of torrefied biomass is generally smaller than the size of raw biomass. How small torrefied biomass particles are depends on torrefaction conditions [46] and intensity of milling. Comparing the value of the biomass char density of table 5.1 and 4.1 we notice that during pyrolysis the torrefied biomass loses about 70% of its weight in the form of volatiles, while raw biomass can lose up to 90%. This difference is due to the torrefaction process where already part of the volatiles, mostly in the form of water vapor, are released and the percentage may vary according to the torrefaction conditions. After the torrefaction, the biomass is lighter and easier to be ground. This yields smaller biomass particles after milling.

For biomass particles, we use the size distribution of torrefied beech wood ‘B-280-5’ from Repellin et al. [46]. This distribution refers to beech chips torrefied during 5 min at 280°C. The Rosin-Rammler distribution which fits these data is shown in
Figure 5.4. Mass loss history for different cut-off diameter of the coal size distribution - 70\(\mu\)m (---), 60\(\mu\)m (----), 50\(\mu\)m (-----), 40\(\mu\)m (⋅⋅⋅⋅), 30\(\mu\)m (-----).

figure 5.11. Its equivalent probability distribution function is shown in figure 5.12. Similar as for coal, the size distribution was cut to avoid simulating the smallest particles. We investigate the effect of the smallest particles on the pyrolysis time by cutting the distribution in the range from 100 to 250\(\mu\)m. For particles smaller than 200\(\mu\)m the criterion to switch from the early stiff phase of pyrolysis to the normal pyrolysis was relaxed to \(r_c = 0.8R\) in order to be able to use the same time step as in chapter 4. The properties of the biomass used are shown in Table 5.1. The mass loss histories in figure 5.13 show that the smallest particles only influence the early phase of pyrolysis, while in the late phase the different lines are very close to each other. The close-up view in the region between 99 and 95\% of the total biomass conversion shown in figure 5.14 leads to the same conclusion as for the coal distribution: no systematic trend of the pyrolysis time with the cut-off diameter can be observed. Therefore, the cut-off for the torrefied biomass particle distribution is set to 150\(\mu\)m. From the equivalent volume distribution function associated with the pdf of biomass particle diameter in figure 5.15 it is visible that a cut-off diameter in this range means that only a small volume of the torrefied biomass is ignored (less than 2\% for a cut-off diameter of 150\(\mu\)m).

In the next section we analyze the co-pyrolysis of coal and biomass and we illustrate the important role played by the biomass particle size and the reason why it is important to have the biomass particle size comparable to the coal particle size.
Figure 5.5. Close-up view of the mass loss history between 95% and 99% of the total coal conversion - 70µm (---), 60µm ( - - - - - - ), 50µm ( --- ), 40µm ( ··········· ), 30µm ( --- ).

Table 5.2. Volume fractions and heating values of coal and biomass used in the simulations.

<table>
<thead>
<tr>
<th></th>
<th>Mass fraction</th>
<th>Volume fraction</th>
<th>Volume fraction</th>
<th>Heating value (MJ/kg)</th>
</tr>
</thead>
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<tr>
<td></td>
<td>stoichiometric</td>
<td>stoichiometric</td>
<td>20% excess</td>
<td></td>
</tr>
<tr>
<td>coal</td>
<td>1/8.806</td>
<td>3.37 × 10⁻⁵</td>
<td>2.88 × 10⁻⁵</td>
<td>25.23</td>
</tr>
<tr>
<td>torrefied biomass</td>
<td>1/6.360</td>
<td>6.10 × 10⁻⁵</td>
<td>5.23 × 10⁻⁵</td>
<td>17.8</td>
</tr>
</tbody>
</table>
Figure 5.6. Close-up view of the mass loss history in the early phase of pyrolysis - 70µm (---), 60µm (---), 50µm (- - -), 40µm (-----), 30µm (----).

Figure 5.7. Volume distribution function associated with the pdf of coal particle diameter.
5.2 Coal and biomass particles size distribution

Figure 5.8. Coal mass loss for different particle factors - factor 3 (······), factor 5 (−−−), factor 10 (---), factor 20 (----), factor 50 (-----), factor 100 (•), factor 200 (×).

Figure 5.9. Close-up view of the mass loss between 95% and 99% of the total coal conversion for different particle factors - factor 3 (······), factor 5 (−−−), factor 10 (---), factor 20 (----), factor 50 (-----), factor 100 (•), factor 200 (×).
Figure 5.10. Coal mass loss for different particle factors and volume fraction equal to $2 \times 2.88 \times 10^{-5}$. Factor 20 (---), factor 50 (--), factor 100 (-----).

Figure 5.11. Cumulative distribution function of torrefied beech wood particle diameter - Rosin-Rammler distribution (---) and experimental data (●) [46].
Figure 5.12. *Probability distribution function of torrefied beech wood particle diameter.*

Figure 5.13. *Mass loss history for different cut-off diameter of the torrefied biomass diameter distribution - 250µm (—), 200µm (-----), 150µm (—-), 100µm (——).*
Figure 5.14. Close-up view of the mass loss history between 95% and 99% of the total torrefied biomass conversion - 250µm (---), 200µm (-----), 150µm (--), 100µm (----).

Figure 5.15. Volume distribution function associated with the pdf of torrefied beech wood particle diameter.
5.3 Co-pyrolysis

In the introduction the synergetic effect of co-pyrolysis for a better utilization of coal was explained. In the present section we do not consider this effect, but consider the effect of blending ratio, pyrolysis temperature and coal particle diameter distribution in co-pyrolysis of coal and biomass on the mass loss history and pyrolysis time.

First, we show the results of the co-pyrolysis simulations with blending ratio ranging from 10% to 50% (energy based) to analyze the dependency of the pyrolysis time on the blending ratio. Typical volume fractions of co-firing are shown in table 5.2.

5.3.1 Varying blending ratio and volume fraction

In our simulations we use the volume fraction based on 20% air excess and since we only simulate pyrolysis, the gas in the simulations is nitrogen.

In figure 5.16 the total mass loss histories are shown as the sum of the coal and biomass particles. The figure shows a substantial dependence of the total mass loss on the blending ratio, which is due to the dependence of the final char yield on the blending ratio.

Analysis of the mass loss histories of the biomass and coal particles separately shown in figure 5.17 indicates that the blending ratio does not have any significant influence on the pyrolysis characteristics of coal. Instead, the pyrolysis time of biomass particles varies with blending ratio, as a close-up view of the late phase of the biomass pyrolysis process shows in figure 5.18. However, the results do not show a systematic trend with blending ratio. The differences are caused by the different particle diameter distribution functions, which are not exactly the same for different blending ratio. We consider this a random effect. Moreover, this results suggest that two-way coupling effects do not play an important role. We investigated this by increasing the volume fraction by a factor of ten.

Figure 5.19 shows the mass loss history for various blending ratios for a situation in which the volume fraction is ten times higher. Such a high volume fraction is unrealistic for combustion applications, since there is too little air for complete combustion of all coal and biomass. However, there are two relevant applications for this volume fraction: pyrolysis and Oxyfuel combustion. Pyrolysis is carried out in a non-reacting gas, so there is no limiting agent from the gas phase to the process. Oxyfuel combustion refers to combustion in air enriched with oxygen to produce a CO\textsubscript{2} rich flue gas ready for sequestration.

Compared to the results at the lower volume fraction in figure 5.16, the ten times higher volume fraction results show a delay of the pyrolysis conversion time. This effect is confirmed by the analysis presented in chapter 4 for raw biomass particles, where the effect of simulating volume fraction was analyzed. In chapter 4 we have discussed the effect of simulating pyrolysis of biomass particles for different particle sizes and volume fractions. There were two conclusions: 1) for the same volume fraction the two-way coupling effect is stronger in the presence of smaller than in the presence of bigger particles; 2) larger volume fractions delay pyrolysis conversion.

Now we discuss the effect of varying blending ratio at low and high volume fraction. A higher blending ratio implies two effects: 1) there are relatively more large
particles since biomass particles are larger than coal particles; 2) the volume fraction is larger, since the heating value of biomass is lower than of coal. The results of chapter 4 imply that these two effects will have an opposite effect on the pyrolysis conversion time. Indeed, this is visible at the higher volume fraction, where a close-up view of the initial phase of pyrolysis in figures 5.20 shows that both for coal and biomass particles a higher blending ratio systematically accelerates the devolatilization. This means that in this initial phase effect 1 dominates over effect 2. On the other hand, in the late phase of pyrolysis in figures 5.21 effect 2 dominates over effect 1. It can be expected that the effects of two-way coupling are proportional to the volume fraction. Therefore, both these systematic two-way coupling effects related to varying blending ratio are absent at the lower volume fraction studied in figure 5.18 and we can conclude that the variations in mass loss history with blending ratio at this lower volume fraction are caused by random effects in the particle size distribution.

![Figure 5.16](image)

**Figure 5.16.** Mass loss of coal and biomass normalized by the initial particle mass for different blending ratios - 10% (---), 20% (---), 30% (---), 40% (-----), 50% (—).
5.3 Co-pyrolysis

Figure 5.17. Mass loss of coal and biomass separately and normalized by their initial mass for different blending ratios - 10% (---), 20% (——), 30% (-----), 40% (------), 50% (-----).

Figure 5.18. Close-up view of the biomass mass loss for different blending ratios - 10% (---), 20% (——), 30% (-----), 40% (------), 50% (-----).
Figure 5.19. Mass loss of coal and biomass separately and normalized by their initial mass with volume fraction 10 times larger - 10% (---), 20% (--), 30% (---), 40% (-----), 50% (——).

Figure 5.20. Close-up view of the coal and biomass mass loss lines in the early phase of pyrolysis for different blending ratios with volume fraction 10 times larger - 10% (---), 20% (---), 30% (--), 40% (-----), 50% (——).
Figure 5.21. Close-up view of biomass mass loss lines in the late phase of pyrolysis for different blending ratios with volume fraction 10 times larger - 10% (---), 20% (---), 30% (--), 50% (- - -).
5.3.2 Varying blending ratio and coal pyrolysis temperature

In this section we show the effect of varying pyrolysis temperature of coal. The pyrolysis temperature of coal $T_{pc}$ is set to 600, 700 and 800 K, while for the biomass it is always kept at 600 K. Typically, coal pyrolysis starts at a higher temperature than biomass pyrolysis. Torrefied biomass also pyrolyzes at higher temperature than raw biomass due to the torrefaction pre-treatment. We analyze the effect of varying $T_{pc}$ for different blending ratios.

The larger pyrolysis time of coal delays coal pyrolysis as figure 5.22 shows. Black, blue and red colors refer to $T_{pc}$ equal to 600, 700 and 800 K, respectively. The close-up view on coal mass loss in the early phase of pyrolysis in figure 5.23 shows that a smaller blending ratio leads to a slower devolatization of coal if the coal pyrolysis temperature is kept fixed (lines with the same color). This illustrates that two-way coupling effect 1 is dominant. A close-up view of the biomass mass loss in the late phase of pyrolysis in figure 5.24 shows that the final pyrolysis time of biomass particles shows a systematic delay with blending ratio: higher blending ratios lead to a longer pyrolysis time due to the associated higher volume fraction. This indicates that again two-way coupling effect 2 is dominant. These results show that for the same $T_{pc}$ the two-way coupling effects of varying blending ratio are similar as in the previous section. Varying the coal pyrolysis temperature does affect the pyrolysis of coal, but does not influence the subsequent biomass pyrolysis, as shown in figure 5.24.

Of the parameters studied here varying the coal pyrolysis temperature has the largest influence. We can conclude that the type of coal, quantified by its pyrolysis temperature, is an important parameter for obtaining comparable devolatization times for biomass and coal. Another parameter that can be varied is the particle size. In the next section the effect of varying the size of coal particles is studied.
5.3 Co-pyrolysis

Figure 5.22. Mass loss of coal and biomass separately and normalized by their initial mass with volume fraction $2.88 \times 10^{-4}$ and for different $T_{pc}$ - coal and biomass lines indicated in the picture with labels - black refers to $T_{pc} = 600$ K, blue to $T_{pc} = 700$ K, and red to $T_{pc} = 800$ K - solid refers to blending ratio 10%, dashed to 30% and dotted to 50%.

Figure 5.23. Close-up view of normalized mass loss of coal in the early phase of pyrolysis with volume fraction $2.88 \times 10^{-4}$ and for different $T_{pc}$ - coal and biomass lines are indicated with labels in the picture - black refers to $T_{pc} = 600$ K, blue to $T_{pc} = 700$ K, and red to $T_{pc} = 800$ K - solid refers to blending ratio 10%, dashed to 30% and dotted to 50%.
Figure 5.24. Close-up view of normalized mass loss of biomass in the late phase of pyrolysis with volume fraction $2.88 \times 10^{-4}$ and for different $T_{pc}$ - black refers to $T_{pc} = 600$ K, blue to $T_{pc} = 700$ K, and red to $T_{pc} = 800$ K - solid refers to blending ratio 10%, dashed to 30% and dotted to 50%.
5.3.3 Varying coal particle diameter distribution

Coal particle diameter distributions are modeled using Rosin-Rammler distribution functions, as described in section 4.8.3. The shape and scale parameters of the coal particle distribution shown in sub-section 5.2.1 is $k = 1.15$ and $\lambda = 0.053$ (see section 4.8.3). In this sub-section we show the effect of larger coal particles on co-pyrolysis by changing the scale parameter $\lambda$. We used two more coal particle diameter distributions with scale parameter $\lambda = 0.1$ and 0.3, shown in figure 5.25.

Larger coal particles need a longer time to pyrolyse, as shown by the mass loss histories in figure 5.26. The blending ratio is 50% and the volume fraction is $2.88 \times 10^{-4}$. The coal particle size distribution affects both coal and biomass devolatization. In the beginning of the pyrolysis process the presence of more smaller coal particles in the distribution with $\lambda = 0.053$ results in a larger transfer of heat from the gas, due to the associated larger coal particle surface area. This results in a delayed devolatization of biomass. In later phases of the process, the gas is heated by the hot walls because of thermal diffusion and the resulting biomass pyrolysis time is smaller than for $\lambda = 0.1$ and 0.3. The relatively comparable size of coal and biomass particles for $\lambda = 0.1$ and 0.3 leads to comparable devolatization times. For these cases the bigger coal particles extract less heat from the gas in the beginning compared to $\lambda = 0.053$. But later, pyrolysis is occurring almost simultaneously, hence the gas is cooled faster and the pyrolysis times becomes larger.

![Figure 5.25](image)

**Figure 5.25.** Probability distribution functions of coal particle diameter - $\lambda = 0.053$ (—), $\lambda = 0.1$ (—), and $\lambda = 0.3$ (---).
5.4 Synergy in co-pyrolysis

Experimental studies [59, 26, 52] report synergetic effects in co-pyrolysis while some other studies do not [43, 6]. The discovery of this synergy is recent and still not clearly understood. The supply of hydrogen from biomass to coal has been addressed as the cause of the synergy [59]. In this section a model for the co-pyrolysis synergy is proposed to only show that our formulation offers the capability to simulate the synergy if a proper model will be developed. The model we propose is based on experimental research reported in literature and it is not meant to explain the synergy.

Our Euler-Lagragian formulation offers the advantage to directly simulate the interaction between gas and particles and to accurately predict particle dispersion. These two characteristics are very important in simulating phenomena that strongly depend on gas-particle interaction and particle dispersion. In case of synergetic effects, accurate prediction of the number of biomass particles in a certain region around a coal particle is fundamental. These advantages are also the motivation for developing our Euler-Lagrangian formulation to simulate co-firing in the future. For the time being, we do not consider combustion and, therefore, we show the advantage in case of co-pyrolysis, with a basic synergy model. When a better synergy model will be developed, our simulation method offers the flexibility to easily implement it.

During pyrolysis, raw biomass can typically lose almost 90% of its weight while torrefied biomass typically loses 70% of its original weight (see section 5.2.2). On the other hand, coal loses typically only 30% of its weight [22]. In table 5.3 the elemental and proximate analysis of the biomass and coal used in the simulations show that the amount of fixed carbon in a coal particle is 57.8% whereas in a biomass particle it is only 21.6%. Since the fixed carbon amount cannot be volatilized, it determines the minimum final weight of the particle after complete pyrolysis. Comparing the volatile
5.4 Synergy in co-pyrolysis

matter of biomass and coal in table 5.3, it is clear that more volatile matter could still be extracted from coal if only the fixed carbon would be left.

Zhang et al. [59] investigated the effect of co-pyrolysis of legume straw biomass and Dayan lignite coal, finding that the amount of extra volatiles obtained from coal depends on blending ratio and temperature. Instead, no synergetic effect was observed for the biomass. Haykiri-Acma and Yaman [26] found that synergy depends also on the coal used. The synergy is typically observed for some blending ratios and temperature ranges. Within these ranges the extra amount of coal volatiles with respect to the amount released if only coal was pyrolyzed may vary. The phenomenon has not been studied in detail, but Zhang et al. [59] attribute the synergetic effect to the larger hydrogen-carbon ratio of biomass compared to coal, i.e. to the extra amount of hydrogen present in the biomass volatiles.

Based on that, we developed a model which takes into account the interaction between the biomass volatiles and the coal particles in order to predict the final char yield of coal as a function of the blending ratio. We did not investigate the effect of temperature because we use the following reaction to compute the composition of the biomass volatiles:

\[ C_nH_mO_k \rightarrow \frac{m}{2} H_2 + k CO + (n - k) C, \]  \hspace{1cm} (5.1)

which holds only for very high temperatures. Therefore, the initial temperature of the channel is always set to 1400 K. It has been shown that at this temperature CO and H\(_2\) are the main components of biomass volatiles [30]. Hence, the pyrolysis process together with the elemental analysis of torrefied wood in Table 5.3 and reaction (5.1) predicts the released mass of hydrogen gas as a function of time.

As a next step in our model, we assume that all hydrogen supplied by the biomass reacts with the char of coal producing methane:

\[ C + 2H_2 \rightarrow CH_4. \]  \hspace{1cm} (5.2)

In order to compute the amount of C volatilized from the char according to (5.2), the

<table>
<thead>
<tr>
<th></th>
<th>Beech wood</th>
<th>Torrefied wood</th>
<th>Bituminous coal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proximate analysis</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Moisture (wt%,wb)</td>
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<td>2</td>
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<tr>
<td>Volatile matter (wt%,db)</td>
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<td>71.5</td>
<td>30.1</td>
</tr>
<tr>
<td>Fixed carbon (wt%,db)</td>
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<td>27.6</td>
<td>57.9</td>
</tr>
<tr>
<td>Ash (wt%,db)</td>
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<td>0.91</td>
<td>12.0</td>
</tr>
<tr>
<td>Elemental analysis (wt%,db)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>47.8</td>
<td>49.9</td>
<td>66.2</td>
</tr>
<tr>
<td>H</td>
<td>6.3</td>
<td>4.9</td>
<td>4.7</td>
</tr>
<tr>
<td>N</td>
<td>0.4</td>
<td>0.17</td>
<td>1.4</td>
</tr>
<tr>
<td>O (by difference)</td>
<td>45.1</td>
<td>44.09</td>
<td>15.7</td>
</tr>
</tbody>
</table>

Table 5.3. Proximate and elemental analysis of wood, torrefied wood and coal [20] - db = dry basis, wb = wet basis.
concentration of hydrogen at the coal particle position needs to be known. However, we did not implement a convection-diffusion-reaction equation for hydrogen gas, but as a first step we considered two simpler models:

- a **global model** in which all hydrogen gas released by the biomass is instantaneously available for the coal particles, independent of their position in the channel.
- a **local model** in which the char of coal can react only with the hydrogen gas supplied by the biomass particles present in the same control volume.

Since only the char of coal can react with hydrogen gas, the amount of char available for the conversion is determined by the position of the char front in the coal particle during pyrolysis. For both models the hydrogen gas which does not take part in the char devolatization is added to the gas phase and can no longer react.

In the global model the hydrogen gas supplied by the biomass is distributed over the coal particles with char available for devolatization. The mass of hydrogen gas available for particle $i$, $H_i$, is proportional to the particle surface area $S_i$:

$$H_i = H_{tot} S_i / \sum_j S_j,$$

where $H_{tot}$ is the total mass of hydrogen gas released by the biomass particles at time $t$, and $\sum_j S_j$ is the sum of the surface area of all coal particles in the channel with char available for devolatization.

In the local model only the hydrogen released by the biomass at time $t$ in the same control volume as the coal particle is available:

$$H_i = H_{CV} S_i / \sum_{j \in CV} S_j,$$

where $H_{CV}$ is the mass of hydrogen in the control volume and $\sum_{j \in CV} S_j$ is the sum of the surface area of the coal particles (with char available for devolatization) present in the same control volume at time $t$.

The disadvantage of the global model is that it does not account for the distance between the particles, which means that hydrogen diffuses at infinite velocity. On the other hand, the local model accounts better for the relative distance and, therefore, for the local hydrogen gas concentration at the time the biomass volatiles are released. However, if a coal and biomass particle are very close to each other but located in neighboring control volumes they will not interact.

Because of the synergy, the mass rate of change of coal particle $i$ is written as the sum of the mass loss due to pyrolysis and the loss of carbon due to synergy with the biomass particles:

$$\frac{dm_i}{dt} = \left. \frac{dm_i}{dt} \right|_{pyr} + \left. \frac{dm_i}{dt} \right|_{syn},$$

(5.3)

where $\left. \frac{dm_i}{dt} \right|_{pyr}$ is given by 4.15. The amount of carbon which devolatiles due to the
Synergy in co-pyrolysis is modeled as follows:

\[
\left. \frac{dm_i}{dt} \right|_{syn} = -C_{syn} \frac{d}{dt} H_i
\]  

(5.4)

where \( C_{syn} \) is the synergy constant. We derived \( C_{syn} \) considering that:

- The mass of hydrogen gas is computed from the mass of volatiles released by the biomass particles at time \( t \) only. From (5.1) it is derived that, in terms of mass, about 4% of the biomass volatiles consists of hydrogen; this provides \( H_{tot} \) or \( H_{CV} \) depending on the model;
- The ratio of carbon mass and hydrogen mass required for reaction (5.2) equals 3.

Therefore, the synergy constant is:

\[ C_{syn} = 0.12. \]

The synergetic effect of co-pyrolysis was investigated using both models and for varying blending ratio in the range 50% to 80%. The difference in mass loss between the two models and the mass loss without synergy was analysed for blending ratios 50% and 80%.

First, we discuss the differences in mass loss for blending ratio 80%. The coal mass loss obtained using the global and local model is compared with the result without synergy in figure 5.27. The biomass mass loss is not compared since it does not show any relevant difference. This is due to the low volume fraction based on stoichiometric conditions. Both models show that synergy leads to a larger devolatization of coal. Using the global model, coal devolatization is very fast. Indeed, the process seems to be suddenly interrupted when the coal mass loss reaches its maximum value allowed, about 41%. On the other hand, the results of the local model show a smoother behavior. In the local model, coal does not devolatize completely, because its rate of devolatization due to synergy \( \left. \frac{dm}{dt} \right|_{syn} \) is limited by the amount of hydrogen gas released by the biomass particles in the same control volume. For blending ratio 50% the two models also show a large difference. However, even for the global method the total hydrogen available is not sufficient to completely devolatize coal particles, as shown in figure 5.28.

Since the local model accounts better for local effects than the global model, we used the local model to compare the results of varying blending ratio with synergy incorporated. The mass loss histories in figure 5.29 show that the extra coal devolatization associated with synergy increases with increasing blending ratio. This is explained by the higher amount of hydrogen gas which can be supplied to the coal particles by the larger amount of biomass at a higher blending ratio.
Figure 5.27. Mass loss $m/m_0$ of coal and biomass for blending ratio 80% and volume fraction $2.88 \times 10^{-5}$ - no synergy (---), global model (---), local model (---), biomass (*)

Figure 5.28. Mass loss $m/m_0$ of coal and biomass for blending ratio 50% and volume fraction $2.88 \times 10^{-5}$ - no synergy (---), global model (---), local model (---), biomass (*)
Figure 5.29. Mass loss $m/m_0$ of coal and biomass for the local synergy model for different blending ratios and volume fraction $2.88 \times 10^{-5}$ - 50% (---), 60% (-- --), 70% (------), 80% (-----)
5.5 Conclusions and recommendations

The biomass pyrolysis model proposed by Haseli et al. [25] has been further developed in order to combine it with DNS of the gas phase. In that way, simulations of biomass particles undergoing pyrolysis have been performed to study the effects of particle-gas interaction on the process.

Direct numerical simulations shows that this interaction leads to pyrolysis times different than the ones predicted using the same pyrolysis model for a single particle. This can be attributed to the fact that the temperature of the gas decreases due to the convective heat transfer with the cooler particles. This coupled effect is taken into account in this formulation and shows that for increasing volume fraction the pyrolysis process is delayed. More particles extract more heat from the gas. However, this effect is only significant for sufficiently high volume fractions. It has been found that the range of volume fractions for which two-way coupling of energy is important agrees with the range of volume fractions for which two-way coupling of momentum is important.

The convective heat transfer coefficient for heat transfer between spherical particles and gas is inversely proportional to the diameter of the particle. Therefore, for the small particles considered in biomass and coal pyrolysis the most important mechanism for heat transfer in the beginning of the pyrolysis process is not radiation but convection. Since the convective heat transfer depends on the local gas temperature, two-way coupling effects by a reduction of the gas temperature are more important for smaller than for larger particles. In particular, the effect of volume fraction and particle size on the pyrolysis conversion time shows that not only the particle volume fraction but also the particle size determines the magnitude of the effect of two-way coupling. For the same volume fraction, smaller particles extract more heat from the gas than bigger particles, due to the larger total surface area and the higher convection heat transfer coefficient. These observations were clearly shown by the results of simulations of co-pyrolysis at high volume fraction, where the two effects due to a higher blending ratio, i.e. higher volume fraction and larger particle size, are combined and counteract each other.

The investigation of biomass pyrolysis using realistic particle diameter distributions and mass fractions based on stoichiometric combustion conditions shows that without source terms arising from combustion, the stoichiometric volume fraction is too small to observe clear effects of two-way coupling on the pyrolysis process. On the other hand, at higher volume fractions, these effects are clearly visible.

Concerning co-pyrolysis, the objective of similar behavior of coal and biomass particles can be achieved by using torrefied biomass and coal particles with similar diameter distribution. Another possibility to achieve this is by using this in combination with a type of coal for which the pyrolysis temperature is large enough to delay coal pyrolysis to the time in which also the biomass pyrolysis occurs.

The synergetic effect of co-pyrolysis was also considered in the model. Since the phenomenon of synergy has not been well understood and a model is not yet available, we modeled the effect of the hydrogen gas released by the biomass particles on the coal in a preliminary way. The availability of better models in the future will enable a more thorough study of this effect.
The code will be further developed to simulate the complete process of co-firing. This requires the implementation of combustion and gasification in the model and in the code. Since the coupling terms, especially in the energy equation, are expected to be very large and fast during combustion, special attention needs to be given to the implementation of these extensions. Probably, it will require the use of special numerical time integration methods for stiff systems of ordinary differential equations.

Several other issues need to be addressed in order to obtain a stable numerical method. The presence of a big particle near the wall may lead to instability because its influence on the gas properties in the control volume where the particle is located may be too high. This presents a problem in particular in the region close to the walls, since the size of the control volume is smallest there. If the particle is larger than the control volume, the two-way coupling terms in the gas equations can be distributed over more control volumes. The phenomenon of turbophoresis which leads to mean transport of particles to the walls will increase this effect, especially since turbophoresis is more prominent for larger particles. By studying the problem in homogeneous isotropic turbulence instead of in turbulent channel flow, this problem can be avoided. Both turbophoresis and local small cells are not present in this flow.

For larger particles also the assumption of point particles becomes questionable. If the particle is larger than the typical length scales of the flow, the expressions used for the drag force and the convective heat transfer between gas and particles are no longer valid and finite-size effects need to be incorporated. Finally, incorporation of the effects of the non-spherical particle shape on the correlations for drag force and convective heat transfer is a useful topic for further research.
Coal and biomass co-pyrolysis in DNS of turbulent channel flow
References

References


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Curriculum vitae

Emanuele Russo was born on August 27, 1983 in Mugnano di Napoli, in Italy. After finishing the scientific liceum in 2001 at Liceo Cartesio in Giugliano in Campania (IT) he studied Aerospace Engineering at the Seconda Università degli Studi di Napoli, in Aversa (IT). During the Bachelor, he spent five months as intern at the Italian Centre of Aerospace Research in Capua, to work on his final project investigating the flow separation on a test-bed. He graduated cum laude in 2005 and then started the Master of Science at the same university. In 2009 he was awarded an Erasmus scholarship to complete part of his Master program at the Technische Universität München in Germany. He graduated cum laude in 2009 and in February 2010 he started a PhD project at the Technische Universität Eindhoven in the Netherlands of which the results are presented in this dissertation. He carried out his research under the supervision of Prof. Hans Kuerten, Prof. Bernard Geurts and Dr. Cees van der Geld. The research focused on the coupling of momentum, mass and heat between the carrier phase and dispersed phase of a two-phase channel flow. The modeling of the dispersed phase is developed in such a way that particles or droplets can be treated as point particles in the flow without losing accuracy in the results. His research was applied to study evaporation and condensation in water droplet-laden turbulent channel flow. Later, biomass particles undergoing pyrolysis were investigated in terms of changes due to the particle-gas interactions in a fully coupled three-dimensional system. Meanwhile he tutored groups of Bachelor students and supervised projects of Master and Bachelor students.

During his studies in Italy Emanuele was working in his family company and was tutor in Mathematics of high and middle school students. In München he worked at Experteer Careers and later as intern at Deutsche Bahn Mobility logistic AG. He played volleyball in several teams. With ASD Olimpia Arzano he shifted from the first division to Serie C of the Italian volleyball league. In 2009 he was European champion in Zürich, with Team München. He is nowadays playing volleyball as a hobby and he started practicing Taekwondo.