Delay of biomass pyrolysis by gas–particle interaction

E. Russo\textsuperscript{a}, J.G.M. Kuerten\textsuperscript{a,b,*}, B.J. Geurts\textsuperscript{b,c}

\textsuperscript{a} Department of Mechanical Engineering, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands
\textsuperscript{b} Multiscale Modeling and Simulation, Faculty EEMCS, University of Twente, P.O. Box 217, 7500 AE Enschede, The Netherlands
\textsuperscript{c} Anisotropic Turbulence, Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

\section*{A R T I C L E   I N F O}

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\section*{A B S T R A C T}

We apply a biomass pyrolysis model, based on the model developed by Haseli et al. [4], which can be used in combination with Direct Numerical Simulation. The pyrolysis model is combined with a model for particle tracking to simulate 3D turbulent particle-laden channel flow with biomass particles undergoing pyrolysis in nitrogen. Transfer of momentum, heat and mass between gas and particles are fully taken into account. The effects of this transfer are analyzed and quantified in terms of the delay in the conversion or pyrolysis time. The delay is shown to depend on the initial volume fraction (number of particles) and on the size of the particles. The two-way coupling effects are relevant at volume fractions \(>10^{-3}\). For a fixed volume fraction, gas–particle interaction induces a delay in the devolatilization, decreasing with increasing particle size. Using this model, we also performed simulations of realistic biomass particle size distributions in order to compare two-way and one-way coupling.

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1. Introduction

Biomass has 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 [1]. 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. For instance, the target of reducing CO\(_2\) emissions from coal-fired power plants can be accomplished with co-firing at high biomass fraction and high oxygen concentration [2,3].

During the pyrolysis of biomass, virgin biomass is converted into char and volatiles are released. The volatile composition, concentration and thermal properties depend on the fuel type, temperature, pressure, heating rate and reaction time [4]. Various models of a single biomass particle undergoing pyrolysis are available in literature. Haseli has presented an overview of the available models in [5]. These models are able to capture the variation of important particle quantities during pyrolysis. 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 and the ignition time or the conversion time of the particles. For simulating biomass pyrolysis with a large number of particles, a simplified biomass pyrolysis model is necessary. Here, we present a biomass pyrolysis model based on the model developed by Haseli et al. [5,6], which has the advantage to be suitable in combination with Direct Numerical Simulation (DNS). We combined the pyrolysis model with a model for particle tracking to simulate 3D turbulent particle-laden channel flow with particles consisting of biomass undergoing pyrolysis and interacting with nitrogen. The turbulent gas flow is solved in an Eulerian framework and the particles in a Lagrangian way. To the best of our knowledge, this is the first 3D Euler–Lagrange formulation of biomass pyrolysis.

The presence of particles in a turbulent gas requires that the flow around each particle is solved. 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 [7–9]. 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 [10,11]. 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 [13–16]. By employing Haseli’s pyrolysis model, we extend this approach to
Nomenclature

\( \alpha_B \)  
thermal diffusivity of virgin biomass (m\(^2\)/s)

\( \alpha_C \)  
thermal diffusivity of char (m\(^2\)/s)

\( \Delta h_p \)  
specific heat of pyrolysis (J/kg)

\( \Delta t \)  
time step (s)

\( \hat{q} \)  
heat flux applied to particle surface (W/m\(^2\))

\( \epsilon \)  
emissivity of particle

\( \mu_g \)  
dynamic viscosity of gas (kg/(m s))

\( \nu \)  
kinematic viscosity of gas (m\(^2\)/s)

\( \phi \)  
particle volume fraction

\( Re_T \)  
frictional Reynolds number

\( \rho_p \)  
particle mass density (kg/m\(^3\))

\( \sigma \)  
Stefan–Boltzmann constant (W/(m\(^2\) K\(^4\)))

\( \tau_p \)  
particle relaxation time (s)

\( \tau_w \)  
wall shear stress (kg/(m s\(^2\)))

\( u \)  
velocity (m/s)

\( v_i \)  
particle velocity (m/s)

\( x_i \)  
particle position (m)

\( C_c \)  
specific heat capacity of char (J/(kg K))

\( C_{vol} \)  
specific heat capacity of volatiles (J/(kg K))

\( \rho_p \)  
particle diameter (m)

\( h \)  
heat transfer coefficient (W/(m\(^2\) K))

\( k_g \)  
thermal conductivity of gas (W/(m K))

\( m_i \)  
particle mass (kg)

\( R \)  
particle radius (m)

\( r \)  
radial coordinate (m)

\( r_c \)  
char front (m)

\( r_t \)  
thermal front (m)

\( T \)  
temperature (K)

\( t \)  
time (s)

\( T_0 \)  
initial particle temperature (K)

\( T_p \)  
pyrolysis temperature (K)

\( T_s \)  
particle surface temperature (K)

\( T_w \)  
wall temperature (K)

\( u_r \)  
friction velocity (m/s)

\( v_c \)  
velocity of char front (m/s)

\( Re_p \)  
particle Reynolds number

\( H \)  
half the channel height (m)

Biomass pyrolysis including the heat and mass exchange between gas and particle. Usually, this approach implies uniform temperature of the particle, whereas in the model presented here, the thermal diffusion inside the particle is taken into account although the particle is modeled as a point.

In this paper the steps necessary to obtain the ordinary differential equations from the work of Haseli et al. [6] and to describe the pyrolysis coupled to the flow and the subsequent steps to enable the coupling of the model to DNS of turbulent gas flow are described. The flow regime in burners is typically turbulent to enhance mixing and, therefore, the combustion efficiency. We test the biomass pyrolysis model in a turbulent channel flow. A real burner geometry was used by Ghenai and Janajreh [17], who performed a 2D simulation of co-firing biomass with coal. Ghenai and Janajreh [17] did not solve the turbulent fluctuations directly but used the RANS equations adopting the \( k-\varepsilon \) turbulence model and a stochastic tracking model to predict particle dispersion. In our formulation the particle–gas interactions are modeled in detail. We restrict to the pyrolysis phase, whose prediction, in case of biomass, is very important for a good prediction of the subsequent combustion phase. Implementation of combustion will be a future extension of this research.

An important parameter in the pyrolysis of biomass is the conversion time, i.e., the time biomass particles need to fully convert to char. Due to the very high content of volatiles in biomass, the concentration of volatiles influences the combustion process. With our model we can compute the volatiles concentration at any time and take into account the effects of particle–gas interactions as well. We focus on the pyrolysis conversion or volatilization and in particular analyze the effect of the two-way coupling on the volatilization. We also simulate the mass loss of a realistic biomass particle size distribution to compare the differences between a two-way and a one-way coupling formulation and investigate under which conditions gas–particle interaction considerably delays biomass pyrolysis.

The structure of the paper is as follows. In the next section Haseli’s model is introduced and the steps to make the model suitable for implementation in DNS are described. In Section 3, the gas model is introduced. Then, the numerical methods used to perform the simulations and the simulation set up are described in Sections 4 and 5, respectively. Section 6 is dedicated to the presentation and discussion of the results.

2. Biomass particle model

In this section, we present the pyrolysis model used to derive the equations for the particle temperature during pyrolysis. Then, the equation of motion used for tracking each single particle in the system is presented.

The biomass type considered in this work is torrefied wood. Torrefaction or drying is a thermo chemical 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. It improves the grindability and the hydrophobicity, very important to store the biomass outside. The energy required for torrefaction is partly compensated by the energy reduction for grinding torrefied wood compared to raw wood. The energy requirement for grinding biomass chips is about 1/6 of the heating value. The torrefaction can reduce this energy up to 90% [18].

After milling, two of the main differences between wood and torrefied wood are the smaller size and the more spherical shape of the torrefied biomass particle [19]. The biomass particle model presented here is based on the assumption of spherical shape of the particle, which is a better assumption for torrefied wood than for the raw material.

2.1. The mathematical model of biomass particle pyrolysis

The biomass pyrolysis model of Haseli et al. [5,6] consists of several phases for modeling the overall process. The pyrolysis process is only one of these phases, which are described in the following.

Consider a spherical particle with a uniform initial temperature. When a positive heat flux \( \bar{q} \) 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. [5,6], immediately after the heat flux is applied, the particle is divided into 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 (Fig. 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. This method of introducing a thermal moving front is
called heat-balance integral method and was introduced by Goodman [20,21]. 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 pyrolysis heating, starts (Fig. 1(b)). The first two phases are different only in the mathematical formulation. The pyrolysis phase starts when the surface reaches the pyrolysis temperature. During this phase (Fig. 1(c)), the char front moves from the surface towards the center of the particle (shrinkage 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. [5] remark that, although the choice of a fixed pyrolysis temperature may be controversial, according to other studies [22] 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. Haseli assumes that during pyrolysis the particle does not shrink, thus the volume integrals involved are performed over a constant volume of the particle. The more severe is the torrefaction pretreatment, the better this assumption holds for the small torrefied biomass particles considered in this paper.

Finally, when the particle is completely converted, the char particle is further heated until it reaches the gas temperature. This phase is called post-pyrolysis heating (Fig. 1(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 (Fig. 2(b)). Therefore, Haseli et al. [5,6] distinguish thermally thin and thermally thick particles. The phases of a thermally thin and a thermally thick particle are illustrated in Figs. 1 and 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.

2.1.1. Heating of virgin biomass

The heat equation in spherical coordinates (assuming spherical symmetry) governing the temperature of the particle in the first stage is:

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

(1)

with \(r\) is the radial coordinate, \(t\) the time, \(T(r, t)\) the temperature, and \(\alpha_\theta\) the (constant) thermal diffusivity of the virgin biomass. In Haseli’s approach [5,6] this partial differential equation is transformed to an ordinary differential equation for the surface temperature of the particle by assuming a parabolic temperature profile:

$$\frac{d}{dr}(qf(r_i)) = 60\alpha_\theta q$$

(2)

with \(f(r_i) = 6R^2 - qRr_i + r_i^2 + r_i^4 + R + r_i^2/R^2\), \(r_i\) the thermal front position, \(R\) the radius of the particle, and \(q\) the external heat flux:

$$q = h(T_s - T_i) + \sigma \epsilon (T_s^4 - T_i^4),$$

(3)

written as the sum of convective and radiative heat flux. The convective heat flux is proportional to the relative temperature \((T_s - T_i)\) between the surrounding gas and the particle surface. The heat-transfer coefficient \(h\) is computed using a correlation from literature that is applicable to spherical particles in the range of Reynolds (Re) and Prandtl (Pr) numbers relevant in our simulations. For forced convection around a sphere, the heat-transfer correlation is chosen as [23]:

$$\frac{hR}{k_g} = 1 + 0.3Re_p^{1/2}Pr^{1/3},$$

(4)

where \(k_g\) is the gas thermal conductivity, \(Re_p = |v - u(x, t)| 2R/v\) is the Reynolds number based on the particle diameter and the relative velocity between the particle and the carrier gas at the particle position, and \(v\) is the kinematic viscosity of the gas.

The radiative flux takes into account the heat flux received by the particle from the walls, whose temperature is indicated by \(T_w\) and is constant. The Stefan–Boltzmann constant and the emissivity are indicated by \(\sigma\) and \(\epsilon\), respectively.
The thermal front position \( r_f \) is expressed in terms of \( T_s \) by evaluating the parabolic temperature profile inside the particle at \( r = R \):
\[
  r_f = R - \frac{2k_b q}{\dot{T}_s} (T_s - T_0)
\]  
(5)

with \( k_b \) the thermal conductivity of the virgin biomass and \( T_0 \) the initial particle temperature. Eq. (2) is the governing equation in the first phase. The unknown in (2) is \( T_s \). Eq. (2) can be written in the final form:
\[
  \frac{d\dot{T}_s}{dr} = \frac{60\alpha_g q - f \frac{\partial \dot{T}_s}{\partial T} - \dot{q} \frac{\partial \dot{T}_s}{\partial T} - \dot{T}_g}{f \frac{\partial \dot{q}}{\partial T} + \dot{q} \frac{\partial \dot{q}}{\partial T} - \dot{T}_g},
\]  
(6)

with \( d\dot{T}_g/dr \) the time derivative of the gas temperature at the particle location.

For times close to the initial time, the solution of this equation is approximately proportional to \( \sqrt{T} \). Therefore, explicit time integration methods are not capable of solving this equation accurately. In order to circumvent this problem, we analyze the solution for small values of \( t \). For small values of \( T_0 \), \( \dot{T}_0 \) and \( \dot{q} \) can be assumed constant. A Taylor expansion of \( f(r) \) for \( T_t \) close to \( t = 0 \) shows that \( f(t) \) is proportional to \( (T_0 - T_0)^2 \). Therefore, we solve a differential equation for \( [T_0 - T_0]^2 \), which equals:
\[
  \frac{d\dot{T}_s}{dT} = \frac{2(\dot{T}_0 - T_0)\left( 60\alpha_g q - f \frac{\partial \dot{T}_s}{\partial T} - \dot{q} \frac{\partial \dot{T}_s}{\partial T} - \dot{T}_g \right)}{f \frac{\partial \dot{q}}{\partial T} + \dot{q} \frac{\partial \dot{q}}{\partial T} - \dot{T}_g}.
\]  
(7)

### 2.1.2. Pre-pyrolysis

The difference with the previous phase is the absence of the thermal front. Following Haseli et al. [5] the governing equation in this phase is:
\[
  \frac{d\dot{T}_s}{dT} = \frac{2\alpha_g \dot{q}}{k_b} - \frac{1}{R} \frac{\partial \dot{T}_s}{\partial r}.
\]  
(8)

which is solved for \( T_s \) by numerically integrating the following equation:
\[
  \frac{d\dot{T}_s}{dr} = \frac{2\alpha_g \dot{q}}{k_b} + \frac{1}{R} \dot{q} \frac{\partial \dot{q}}{\partial T} - \frac{\partial \dot{T}_s}{\partial T},
\]  
(9)

A detailed derivation of these equations is given in [24].

### 2.1.3. Pyrolysis

In the pyrolysis phase, the particle is divided into 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 the model the temperature profiles in each region are assumed to be quadratic functions:
\[
  T(r) = \frac{\phi_1}{2}(r - \frac{r_c}{2})^2 + \phi_1(r - r_c) + \phi_0, \quad \text{for} \quad 0 \leq r \leq r_c \quad \text{(10)}
\]
\[
  T(r) = \frac{\phi_2}{2}(r - \frac{r_c}{2})^2 + \frac{\phi_1}{2}(r - r_c) + \phi_0, \quad \text{for} \quad r_c \leq r \leq R. \quad \text{(11)}
\]

The heat equations governing this problem are Eq. (1) for \( 0 \leq r \leq r_c \) and:
\[
  \frac{\partial T}{\partial t} = \alpha_c \frac{1}{R^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) + (1 - \omega) \frac{m}{4r^2} \alpha_{vol} \frac{\partial T}{\partial r}, \quad \text{for} \quad r_c \leq r \leq R, \quad \text{(12)}
\]

with \( \alpha_c \) the thermal diffusivity of the char. The second term on the right hand side of (12) represents heat transfer between the volatiles released at the char front and the char during their transport towards the particle surface. In (12) \( \alpha_{vol} = C_{vol}/\rho_c C_c \) and \( \omega = \rho_c/\rho_B \), with \( C_{vol} \) and \( C_c \) the volatiles and char specific heat capacities, respectively. This term is not considered by Haseli in [5,6] but it has been taken into account in [25].

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 the moving char front and the co-existence of these two regions, in the pyrolysis phase there are three governing ordinary differential equations, whereas in the other phases there is only one. The first governing equation is obtained from energy conservation in the char region and reads as follows:
\[
  \frac{d\dot{T}_s}{dr_c} = \frac{60\alpha_c \dot{q} (r^2 + k_B \dot{q} \phi_1 - r_c^2 v_c \Delta\phi \rho_B + \frac{(1 - \omega)\rho_B v_c C_{vol} C_c^2 (T_s - T_p))}{f \frac{\partial \dot{q}}{\partial T} + \dot{q} \frac{\partial \dot{q}}{\partial T} - \dot{T}_g}.
\]  
(13)

with \( f(r_c, \dot{q}, \phi, \phi_1) = 9R^2a + 6R^2\dot{q} - 11R^2\Delta r_c - 9R^2\Delta r_c + R^2\Delta r_c - R^2\Delta r_c - R^2\Delta r_c - R\Delta r_c + 4\Delta r_c^2 + \Delta r_c^2, \quad a = v_c \rho_B \Delta h_p - k_B \phi_1, \quad \text{with} \quad v_c \text{ the velocity of the char front, and} \quad \Delta h_p \text{ the specific heat of pyrolysis.}

Similarly, the energy conservation in the virgin biomass region yields the second governing equation in the pyrolysis phase:
\[
  \frac{d\dot{T}_s}{dr_c} = -\alpha_{vol} \frac{\partial T}{\partial r_c}. \quad \text{(14)}
\]

Finally, the third governing equation is the definition of the char front velocity:
\[
  \frac{d\dot{r}_c}{dr_c} = v_c. \quad \text{(15)}
\]

The unknowns in the three governing equations are \( T_s, \dot{q}, \phi \), and \( \phi_1 \). The governing equations is written in matrix form. This set of ordinary differential equations are numerically integrated in time [24].

### 2.1.4. Post-pyrolysis heating

This phase is similar to the pre-pyrolysis phase with biomass properties replaced by char properties. Hence, the governing equation is (9) with \( \alpha_B \) and \( k_B \) replaced by \( \alpha_C \) and \( k_C \), respectively.

### 2.1.5. Thermally thick particle

The temperature profile inside a thermally thick particle consists of three regions (see Fig. 2(b)): \( T(r) = T_0 \) for \( 0 \leq r \leq r_t \) \( T(r) = \phi_2(r - r_t)^2 + \phi_1(r - r_c) + \phi_0 \), for \( r_c \leq r \leq r_t \) \( T(r) = \psi_2(r - r_t)^2 + \psi_1(r - r_c) + \psi_0 \), for \( r_c \leq r \leq R \), \( \phi \), whose coefficients have to be determined using the boundary conditions. The energy conservation in the virgin biomass region \( r_t \leq r \leq R \) leads to the first governing equation in this phase:
\[
  \frac{d\dot{T}_s}{dr} = \frac{(R^2 - r_t^2)}{k_B \Delta h_p \rho_B} \left( R^2 - r_t^2 \right) \left( \rho_B \Delta h_p \rho_B + 2k_B \frac{T_p - T_0}{R_c - r_t} \right) \right), \quad \text{(20)}
\]

with \( h(t_r, r_c) = 4r_t^2 + 3r_t^2 + 2r_t^2 + r_t^2 \). The energy conservation in the char region \( r_c \leq r \leq R \) leads to the second governing equation:
\[
  \frac{d\dot{T}_s}{dr} = \frac{(R^2 - r_t^2)}{k_B \Delta h_p \rho_B} \left( R^2 - r_t^2 \right) \left( \rho_B \Delta h_p \rho_B + 2k_B \frac{T_p - T_0}{R_c - r_t} \right) \right), \quad \text{(20)}
\]

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. The stiffness problem can be circumvented by analysis of the problem for very small char and virgin biomass regions (see 3.3 in [24]). By applying approximations based on this analysis in the early and late phases of pyrolysis, the pyrolysis phase is divided into three sub-phases: early stiff pyrolysis sub-phase, regular pyrolysis sub-phase, and late stiff pyrolysis 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 can be derived in the same way as before by assuming a linear temperature profile in the char region in the early pyrolysis sub-phase and constant temperature profile in the virgin biomass region in the late pyrolysis sub-phase.

2.3. Biomass particle routes undergoing pyrolysis

We can 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 Fig. 3. The scheme also shows the criteria used to switch between the phases. By $\varepsilon_R$ 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 $r_t < \varepsilon_R$. Secondly, when $T_s = T_p$ to the early stiff phase of pyrolysis and next, to the pyrolysis phase if $r_c \leq \theta R$, with a typical value

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**Fig. 3.** Scheme of the routes of a biomass particle undergoing pyrolysis – in the flow diagram the criterion which is satisfied first dictates the route followed by the particle.
of \( \theta = 0.9 \). 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 stage when \( T_i = T_p \). Then, if \( r_t \leq \epsilon_k \) before \( r_t \leq 6\epsilon_k \), the thick particle moves to the early stage 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 \( r_t \leq \epsilon_k \) to the pyrolysis phase. Next, it follows the same route as a thin particle.

In the following, the application of this biomass pyrolysis model in DNS is presented.

2.4. Particle equation of motion

We adopt the formulations for drag force on a particle and heat transfer between particle and gas for spherical particles. The same assumption is used by Papadakis et al. [26] and Ma et al. [27], who introduced a shape factor. Incorporation of the effects of a non-spherical particle shape on the correlations for drag force and convective heat transfer is a topic of future research.

Since particles are small and have a much higher mass density than the gas, the drag force and the gravity are the dominant forces exerted on a particle [28]. We do not take gravity into account since settling of particles is not a focal point in this study. Therefore, Newton's law applied to a particle can be written as:

\[
\frac{d(m_i\mathbf{v}_i)}{dt} = m_i\left(\mathbf{u}(\mathbf{x}_i, t) - \mathbf{v}_i\right)\left(1 + 0.15\text{Re}_p^{0.667}\right) + \mathbf{v}_i \frac{dm_i}{dt}. \tag{21}
\]

Here, \( m_i \) is the mass of particle \( i \), \( \mathbf{v}_i \) its velocity and \( \text{Re}_p \) the particle relaxation time given by \( \text{Re}_p = \rho_p d_p^2/(18\mu_g) \). Moreover, \( d_p \) is the particle diameter, \( \rho_p \) is the mass density of the particle and \( \mu_g \) the dynamic viscosity of the gas. The location of the particle is denoted by \( \mathbf{x}_i \) and \( \mathbf{u}(\mathbf{x}_i, t) \) is the velocity of the gas at \( \mathbf{x}_i \). The two terms on the right-hand side are the drag force, where the standard Schiller-Naumann drag correlation valid for particle Reynolds numbers between 0 and 1000 is adopted [29], and the change in momentum due to phase change. Eq. (21) can also be written in terms of accelerations, yielding the following particle equation of motion:

\[
\frac{d\mathbf{v}_i}{dt} = \left(\mathbf{u}(\mathbf{x}_i, t) - \mathbf{v}_i\right)\left(1 + 0.15\text{Re}_p^{0.667}\right) \frac{1}{\text{Re}_p}. \tag{22}
\]

The Lagrangian particle tracking requires integration of the trajectory equation:

\[
\frac{d\mathbf{x}(t)}{dt} = \mathbf{v}_i, \tag{23}
\]

in which the particle velocity \( \mathbf{v}_i \) is computed from (22).

3. Gas model

The computational gas 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. The dispersed phase (particles) is coupled to the carrier phase (gas) by means of source terms reflecting the presence of particles. The coupling between the phases can be classified into three types: (1) "one-way coupling" if only the flow influences the particles; (2) "two-way coupling" if particles have 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 [10]. We adopt two-way coupling, which involves momentum, mass and heat exchange between gas and particles. 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. A model for the volatiles composition is not required in this work since only pyrolysis is considered.

The source terms in the gas equations resulting from two-way coupling satisfy the requirement that they only transfer mass, momentum and internal energy from one phase to the other. They can be written as:

\[
\int_{V} L_{\text{two-way}} dV = - \sum_{i = V} \frac{d}{dt} \begin{bmatrix} m_i u_i \\ m_i v_i \\ m_i w_i \\ m_i \\ \epsilon_i \end{bmatrix}, \tag{24}
\]

where the sum is taken over all particles within volume \( V \). The first three components are the momentum exchange between the two phases, expressed by (21). The fourth component is the mass given by the particle to the gas during pyrolysis in the form of volatiles. The mass rate of change or mass loss is:

\[
\frac{dm_i}{dt} = 4\pi (\rho_B - \rho_C) r_t^2 \nu_c. \tag{25}
\]

In our model, the volatiles added to the gas phase cause an increase in the gas mass density and, consequently, in the pressure, since the volume of our domain is constant and periodic boundary conditions are applied in the streamwise and spanwise directions. For the simulations with the highest mass load of biomass considered here, the resulting increase in pressure is almost 50%. The fifth component is the two-way coupling term of the energy, which is the sum of three contributions:

\[
\frac{d}{dt} \epsilon_i = h A_i (T_g - T_i) + c_{p,g} \frac{dm_i}{dt} - \frac{d}{dt} \left( \frac{1}{2} m_i \mathbf{v}_i^2 \right). \tag{26}
\]

The first term is the convective heat transfer between gas and particle, which is proportional to the temperature difference, the particle surface area \( A_i \) and the convective heat transfer coefficient as defined by Bird et al. [23]. The second term is the energy associated with the volatiles given to the gas by the particle during pyrolysis. The volatiles are formed at the char front where the temperature is \( T_p \). During their traveling to the particle surface, their increase in temperature is negligible. Therefore, to save computational time, the temperature of the released volatiles is set to \( T_p \). In this work, the specific heat capacity of the volatiles and gas is taken equal. The third term is the change in kinetic energy which is expressed by the drag force and the mass loss of the particle:

\[
\frac{d}{dt} \left( \frac{1}{2} m_i \mathbf{v}_i^2 \right) = m_i \frac{dm_i}{dt} \frac{1}{2} \mathbf{v}_i^2 - \frac{1}{2} \frac{dm_i}{dt} \frac{d}{dt} \mathbf{v}_i, \tag{27}
\]

in which \( dm_i/dt \) is given by (22).

4. Geometry and numerical method

The computational domain is a channel with size \( 4\pi H \) in streamwise direction and \( 2\pi H \) in spanwise direction, where \( H = 0.2 \) m is half the channel height (see Fig. 4). 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. The flow solver is described in detail in [30]. The spatial discretization is based on a finite volume method with second-order accuracy.

Due to the coupling terms, it is important that the particle and the gas quantities are evaluated at the same time level. This is obtained using a low-storage second-order accurate Runge–Kutta
method for both gas and particle equations. The time step $\Delta t$ is chosen small enough to ensure a stable and accurate solution of the equations.

In the finite volume method for the gas, the two-way coupling terms (24) account for all particles present in the cell. The quantities in (24) are already computed in the particle equations, which simplifies the implementation of these terms. Details on how to numerically treat the coupling terms are described in [30,24].

5. Set up of the simulations

Simulations are performed at a frictional Reynolds number approximately equal to $Re_f = 150$, based on friction velocity $u_f = \sqrt{\tau_w/\mu}$ and half the channel height, where $\tau_w = \mu \left. \frac{\partial u}{\partial y} \right|_{y=\delta H}$ is the wall shear stress and $u$ the mean streamwise velocity component, averaged over the two homogeneous directions and time. The grid resolution is fine enough for capturing the main elements of the flow characteristics without employing a turbulence model. The initial volume fraction varies from simulation to simulation but it is always smaller than $0(10^{-3})$, so that particle–particle collisions can be ignored but two-way coupling might be relevant [10]. In all simulations shown here the particle size is small compared to the smallest scales of turbulence, quantified by the Kolmogorov length, throughout the channel. Therefore, the point-particle approach employed in this work, is justified.

In Table 1 the properties of the virgin biomass and the char are shown. The virgin biomass density is about 3.4 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 $\sigma = 5.67 \times 10^{-8} \text{W/(m}^2\text{K}^4)$ and $\epsilon = 0.9$. The value of the specific heat of pyrolysis is $\Delta h_p = -270 \text{kJ/kg}$.

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 [31]. 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 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.

6. 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 heat the particles from their initial temperature to the pyrolysis temperature. In the results shown in this section, the conversion time is evaluated at 99% conversion of virgin biomass matter. Results evaluated at 95% of conversion are qualitatively the same.

In Sections 6.2 and 6.3, we investigate the effect of varying volume fraction and particle size on the conversion time. In Section 6.4, we compare the results of the simulations with one-way and two-coupling for a realistic biomass particle size distribution.

6.1. Effects of two-way coupling

The effect of two-way coupling can be observed by studying the time evolution of the temperature profiles of nitrogen and particles. In Fig. 7 the mean nitrogen temperature and mean particle surface temperature are shown as functions of time. The volume fraction $\phi$ is $5 \times 10^{-2}$. The particle diameter is 0.4 mm. Initially, the gas is much warmer than the particles, so that the particles receive energy from the gas and from the walls. The volume fraction is very small and the gas temperature is only slightly affected by the presence of the colder particles. The particle temperature rises until it reaches the gas temperature. Afterwards, the particle temperature becomes a little higher than the gas temperature due to the radiative heat flux from the walls, whose temperature is assumed constant (at 1400 K). From these results we conclude that the number of particles in this simulation is too small to appreciate two-coupling effects. Thus, for computing the conversion time of the biomass particle, a constant gas temperature could be a good approximation without requiring the employment of a numerical solver for the gas phase.

A different scenario is presented in Fig. 8. The nitrogen and particle surface mean temperatures are averaged in the homogeneous directions and plotted as functions of the wall-normal coordinate at different instants of time. In this simulation the diameter is the same as above but the volume fraction $\phi$ is $2.55 \times 10^{-4}$ and two-way coupling effects cannot be neglected. In fact, the gas starts to cool down, in the center of the channel more than near the walls (the walls are kept at constant temperature). After a certain time (around $t=0.17\text{s}$), 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 moves 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. Indeed, 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 particle temperature equals the wall temperature.

In Fig. 5, isolines of the gas temperature in a vertical plane of the channel are shown during the initial heating phase of the particles. In the two-way coupling formulation (above), the particles can be identified at the colder spots with lighter colors. In the one-way coupling formulation the particles do not affect the temperature field (figure below). Isolines of temperature during pyrolysis in Fig. 6 show the decrease in gas temperature due to the particles in the two-way coupling formulation, whereas in the one-way coupling formulation the temperature field is not significantly changed.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Properties of the virgin biomass and the biomass char used in the simulations.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mass density (kg/m$^3$)</td>
</tr>
<tr>
<td>Biomass</td>
<td>650</td>
</tr>
<tr>
<td>Char</td>
<td>190</td>
</tr>
</tbody>
</table>
We performed simulations with different numbers of particles in order to analyze the dependence of the pyrolysis conversion time on the particle volume fraction.

6.2. Dependence on volume fraction

The diameter of all particles was 0.4 mm. The results in Fig. 9 show that in case two-way coupling is applied the conversion time increases with increasing volume fraction and it is constant for 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 Fig. 9 shows that the particle feedback to the gas becomes important for accurately predicting the conversion time when $\phi > 10^{-5}$. The conversion time for $\phi = 10^{-3}$ is twice the conversion with one-way coupling, whereas for $\phi < 10^{-4}$ the two-way coupling only leads to negligibly small differences. The range of volume fractions in which two-way coupling for the energy is relevant is equal to the range in which two-way coupling of momentum is important [10]. According to Elghobashi [10], for $\phi > 10^{-3}$ four-way coupling of momentum needs to be considered. We limited our investigation to two-way coupling so we cannot say whether or not the four-way coupling for the energy (radiation between the particles) is also important if $\phi > 10^{-3}$.

6.3. Dependence on particle size

In order to investigate the effect of particle size on the conversion time, we performed simulations at constant volume fraction and varying particle diameter. Fig. 10 shows the conversion time $t_{\text{conv}}$ dependence on the particle diameter for three different volume fractions: low, medium and high. For each volume fraction,
Fig. 6. Pyrolysis phase: instantaneous gas temperature in Kelvin in a vertical plane of the channel for the simulation with $\phi = 2.55 \times 10^{-4}$ and $d_p = 0.4$ mm; above: two-way coupling formulation; below: one-way coupling formulation.

$t_{\text{conv}}$ increases with increasing diameter. For the same diameter, the conversion time $t_{\text{conv}}$ at a higher $\phi$ increases more compared to a lower $\phi$ or one-way coupling. The effect of two-way coupling is more clear using a logarithmic scale in which it is visible that, depending on the volume fraction, $t_{\text{conv}}$ is proportional to a certain power of $d_p$, whereas for the one-way coupling formulation (or very low $\phi$) $t_{\text{conv}} \propto R^{5/3}$. The figure also shows 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 can be 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. (4)).

According to Fig. 8, the delay in conversion time due to two-way coupling strongly depends on $\phi$ but seems to be approximately independent of $d_p$. These results suggest that, in first
approximation, the delay in the conversion time for a given diameter and volume fraction can be estimated from the conversion time computed with a one-way coupling formulation, which is much less computationally expensive.

6.4. Particle size distribution

In practical applications of co-firing of biomass and coal, the probability density function of biomass particle diameter follows the Rosin–Rammler distribution [32]. This function $P(d_p)$ is defined as follows:

$$P(d_p) = \frac{k}{\lambda} \left( \frac{d_p}{\lambda} \right)^{k-1} e^{-(d_p/\lambda)^k}$$

with $k$ and $\lambda$ the shape and scale factors, respectively.

Here, we consider the size distribution of torrefied beech wood ‘B–280-S’ from Repellin et al. [18]. This distribution refers to beech chips torrefied for 5 min at 280°C. The Rosin–Rammler distribution which fits these data is shown in Fig. 11, with $k = 1.17$, $\lambda = 0.18$ and the diameter in millimeter. The size of torrefied biomass is generally smaller than the size of raw biomass. How small torrefied biomass particles depends on torrefaction conditions and intensity of milling [18]. After the torrefaction, the biomass is lighter and easier to be ground. This yields smaller biomass particles after milling.

The presence of very small particles requires a very small time step because of the stability restriction associated with the explicit time-integration method used here. Including very small

Fig. 8. Mean gas temperature (--) and particle surface temperature (---) at different instants of time for an high volume fraction ($\phi = 2.55 \times 10^{-4}$).

Fig. 9. Conversion time at different volume fractions with particle diameter 0.4 mm and initial nitrogen temperature 1400 K; one-way coupling (x); two-way coupling (●).
particles would lead to very long computational times. Therefore, we bounded the size distribution from below, in such a way that very small particles are disregarded without noticeably influencing the results [24]. This saves computing time.

In order to study the importance of gas–particle interaction on the pyrolysis conversion time, we compare the particle mass histories of the two-way coupling simulation and a simulation without two-way coupling. The comparison in Fig. 12 suggests that two-way coupling effects are of minor importance due to the low particle volume fraction used in these simulations. The delay is approximately

8% compared to the one-way coupling simulation. Indeed, the volume fraction is $5.23 \times 10^{-5}$, 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) [31].

However, in our model combustion is not modeled. Therefore, the amount of biomass can be increased. Moreover, also with combustion in Oxyfuel conditions, where the oxygen concentration is very high, the biomass ratio can be higher than the stoichiometric value. 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 more than 50% with respect to the same simulation with one-way coupling (Fig. 13).

7. Conclusion

In this paper we have shown how to extend Haseli’s pyrolysis model for biomass to an Euler–Lagrange formulation which makes the model suitable for implementation in a DNS code. Our 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 biomass pyrolysis that may strongly depend on gas–particle interaction and particle dispersion, as we have shown in the results. In particular, the heat and mass exchange between particles and gas influences the biomass pyrolysis conversion. The conversion time increases with increasing volume fraction and particle size. The particle feedback to the gas is only important...
if $\phi > 10^{-5}$. The conversion time delay is proportional to a certain power of the particle diameter depending on the volume fraction. In first approximation, the delay can be estimated from the one-way coupling result, thus saving computational power. Our formulation allows the simulation of a realistic biomass size distribution for predicting the biomass loss history. Theoretically, any size distribution could be simulated and, based on the volume fraction, the particle–gas interactions are also taken into account. For the distribution of torrefied biomass considered in this work, it was found that the very small particles have a minor effect on the conversion time. Therefore, particles with a diameter smaller than 0.2 mm can be ignored.

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