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Development of a transient CFD model for charcoal production in a batch reactor and validation with measurement data



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ARTICLEINFO	ABSTRACT
Keywords: Computational fluid dynamics (CFD) Modelling Charcoal Batch reactor Pyrolysis Biomass	In order to facilitate the development and optimization of a batch reactor for charcoal production, a new CFD model was developed that allows a detailed description of the pyrolysis process of the biomass within the batch reactor. Focusing on computational efficiency, the model is not based on single biomass particles, but on a description of the (local) properties of the packed bed inside the reactor. The drying and pyrolysis processes are described on a local scale, such that according to the prevailing conditions (temperatures, flow) the progress of the reactions can be analysed. To validate the CFD model, dedicated test runs were performed using a pilot-scale pyrolysis reactor, and the measurement results were compared to the simulation. The results show that the progress of conversion is well represented, and a reasonable agreement of temperature profiles inside the retort is achieved. The overall trends for the profile of mass release are also reproduced.

1. Introduction and objectives

Biochar has a broad spectrum of applications, ranging from the production of activated charcoal, over animal feed supplements, pharmaceutical and industrial applications, to the usage in soil improvements. Therefore, in Austria, Germany, and the whole EU, there is a considerable demand of biochar, which is expected to rise in the next decade, due to the change from fossil to renewable raw materials. Currently, most of the charcoal utilized in Austria and Germany is imported either from European countries (Poland, Serbia, Bosnia-Herzegovina) or from Africa and South America, even though raw materials for local production are available (data from "Statistik Austria" and "Statistisches Bundesamt"). Furthermore, the current technology for charcoal production is mostly inefficient and has a high emission intensity.

Therefore, BIOS and Polytechnik Luft-und Feuerungstechnik GmbH are working on the development of a new technology for the fuelflexible, low-emission high-efficiency competitive production of highquality charcoal. At the same time, the process is able to integrate the production of electricity and heat from biomass. The new technology, which has already been patented [1], combines continuous and discontinuous process steps in a very efficient manner, to exploit the advantages of both modes of operation. Fig. 1 shows an overall scheme of the process.

In the production process, the pyrolysis reactor is filled with dried biomass (step 1 in Fig. 1). During pyrolysis operation (step 2), the reactor is heated from the outside with hot flue gas from a gas burner, which burns the generated pyrolysis gas (step 3 in Fig. 1). After the batch duration, the charcoal is extracted from the reactor. In order to achieve an efficient coupling of the discontinuous processes of drying and pyrolysis with the continuous processes of the combustion of the pyrolysis gas, and the heat and electricity generation, the output of several batch reactors in different stages of the pyrolysis process is combined. In this way, the overall efficiency can be maximized.

The heart of the process is the pyrolysis step, and the development and optimization of the pyrolysis reactor is one main point within the project. Of special importance are the influence of the reactor geometry on the progress of the pyrolysis and the temperature profiles inside the reactor. To analyse these aspects, and perform appropriate sensitivity analyses for this large-scale system, a reliable modelling approach is needed. It needs to describe the details of the charcoal production process in the reactor over time in a three-dimensional setting, and at the same time be numerically efficient and stable to allow for the timely generation of results.

Up to now, most modelling approaches for pyrolysis start from a microscopic point of view. Extensive analyses exist concerning the

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Fig. 1. Schematic of the combined process of charcoal, electricity and heat production from biomass.

detailed kinetics of the pyrolysis of biomass [2–6], which have been successfully applied in the context of the pyrolysis of single particles [7]. These models, however, do not consider the interactions of the fuel particles with the gas phase, let alone the effects of a packed bed. Furthermore, they use vast amounts of computational resources, and are therefore not applicable to the simulation of larger systems, such as a large-scale industrial batch reactor. Thus, a new model was developed for the use in this project. As a basis, computational fluid dynamics (CFD) methods are used, which are augmented with a new CFD model specifically developed for the task to describe the charcoal production process in the batch reactor over the whole process duration.

This work focuses on the details of this new model, and its validation. The CFD model was validated using experimental results from a test reactor, where dedicated measurements were performed to investigate not only the batch duration, but also temperature profiles, pyrolysis gas composition and time dependence of the mass release. In addition, several tests were performed in which the batch process was interrupted at different stages, to monitor the carbonization process.

This paper is organized as follows: In section 2, the geometry of the test reactor and the batch operation are described. Section 3 details the newly developed CFD model. Section 4 concerns the test runs used for validating the model, and gives the operating conditions as well as the corresponding parameters used in the simulation. The simulation results are presented in section 5, with a detailed comparison to the experimental findings. Finally, section 6 gives summary and conclusions.

2. Reactor geometry and operation

The test reactor used for validation of the CFD results is a small-scale version of the final reactor used in the production process. Its geometry and layout are shown in Fig. 2.

The innermost part of the reactor is a removable cylindrical retort, which is filled with the biomass. The pyrolysis gas released during the process flows through the packed bed, and through openings at the bottom of the retort into a surrounding channel. From there, it leaves the reactor via a pipe. Outside the channel for the pyrolysis gas, there is another channel for the flue gas utilized to heat the retort. It is supplied via a pipe at the bottom of the reactor and flows around the channel filled with pyrolysis gas. The outlet for the flue gas is located above the inlet, as can be seen in Fig. 2. After the batch is complete, the retort is removed from the reactor, and (after a cooling period) the charcoal is weighed and taken out of the retort.

3. Description of the CFD model

3.1. General approach

Two different routes may be chosen to model a packed bed, such as the one present in the reactor described in section 2: A particle-based approach or a volume-based approach. In the first setup, the individual fuel particles that make up the packed bed are considered separately, and the thermal conversion is tied to their paths through the simulation domain. This type of model was used, e.g., in Ref. [8] to describe the combustion of wood pellets in an underfeed stoker. While providing an accurate description of the processes occurring in the fuel bed, this approach is computationally very demanding. In contrast, the second setup focuses on the properties of the packed bed itself instead of the individual fuel particles. The description of the processes of drying and pyrolysis occurring during the batch is based on the local properties in the fuel bed (e.g., temperature, void fraction of the packing, flow velocity, ...), allowing for different conversion rates in different parts of the bed. This type of model (see also the detailed description in Ref. [9]) is computationally more efficient and easier to implement than the first approach and was therefore chosen for this work. Nevertheless, the interactions of the gas phase and the fuel particles of the packed bed are modelled in detail, especially the distribution of energy among the gas phase and the solid material via convection and radiation. This ensures a good description of the temperature profiles, which are important to model the progress of the pyrolysis.

The model was realised within the commercial CFD software ANSYS Fluent. The additional functionality to describe the batch reactor was implemented using so-called "user-defined functions" in combination with user defined scalar transport equations for the various quantities



Fig. 2. Geometry of the pyrolysis test reactor (top: 3D view, bottom: section through the axis of the cylindrical retort). Explanation: The pyrolysis process is patent protected.

present in the model, as described below.

3.2. Properties of the packed bed

As is the standard in CFD applications, the fuel bed is discretised, along with the rest of the computational domain, into small volumes referred to as "(computational) cells", each of which contains a certain amount of fuel (solid phase) and gas, according to the local porosity (void fraction of the packed bed). For each cell in the fuel bed, the properties of a fuel particle representative for this location are calculated. These are:

- Particle composition (mass of moisture, dry wood, and charcoal in the particle)
- Particle diameter *d_P* (note that the representative particles are considered as spheres)
- Particle density ρ_P
- Particle thermal conductivity λ_P
- Particle heat capacity $c_p^P(T_S)$ as a function of the packed bed temperature T_S

The particle properties change as the process evolves: The particle shrinks as it loses mass via pyrolysis, the density, thermal conductivity, and heat capacity change with the composition.

The properties (thermal conductivity, density, heat capacity) of the particle components moisture, dry wood and charcoal (abbreviated as H_2O,DW,CH) are input parameters of the model (cf. section 4.3), and are used to calculate the particle properties. The heat capacity c_p^P of the particle is the mass weighted average of the components' c_p , as is the thermal conductivity of the wet biomass (dry wood and water). The thermal conductivity of the mixture of dry wood and charcoal (during the stage of pyrolysis) is calculated according to the volume fractions of the respective components. The volumes occupied by the components inside the particle are calculated as

$$V_i = \frac{m_i}{\rho_i} \tag{1}$$

with m_i the mass and ρ_i the density of the component *i*. It is assumed that the moisture present in the particle does not occupy additional volume but fills the wood structure.

The particle volume is the sum of the volumes of the components,

$$V_P = V_{DW} + V_{CH}.$$
 (2)

From the volume of the spherical particle, the particle diameter is obtained as

$$d_P = \sqrt[3]{\frac{6}{\pi}V_P}.$$
(3)

During pyrolysis, as dry wood is converted to charcoal, mass is released from the particle to the surrounding gas phase. From equations (1)–(3) it follows that the approach presented here provides a consistent description of particle shrinkage. Bed compaction is not considered. Therefore, the shrinkage of the particles causes an increase of the porosity of the packed bed: The change over one time step (from ε_0 at the start of the time step Δt to $\varepsilon_0 + \Delta \varepsilon$ at the end of the time step) is

$$\Delta \varepsilon = (1 - \varepsilon_0) \cdot \left(1 - \frac{V_{P,1}}{V_{P,0}} \right)$$
(4)

where $V_{P,1}$ represents the particle volume after the time step, and $V_{P,0}$ the particle volume before the time step.

The pressure loss Δp of the packed bed (divided by the distance *L*) is described with the Ergun equation, see, e.g. [10],

$$\frac{\Delta p}{L} = 150 \ U_0 \frac{\mu_g}{d_p^2} \frac{(1-\varepsilon)^2}{\varepsilon^3} + 1.75 \ U_0^2 \frac{\rho_g}{d_p} \frac{1-\varepsilon}{\varepsilon^3}$$
(5)

with the superficial velocity U_0 , the viscosity of the gas μ_g , and the gas density ρ_g . Note that this description naturally depends on the porosity of the packed bed, and therefore allows for variations of the pressure loss due to particle shrinkage.

The packed bed, described on one hand by the representative particle in each cell and by the local porosity ε on the other hand, is not in thermal equilibrium with the gas flow. The packed bed temperature T_S is calculated via a scalar transport equation (given in cartesian coordinates)

$$\frac{\partial \left(\rho_{S} \int_{T_{ref}}^{T_{S}} c_{p}^{P}(T) dT\right)}{\partial t} - \frac{\partial}{\partial x_{i}} \left(\lambda_{S} \frac{\partial T_{S}}{\partial x_{i}}\right) = \dot{Q}_{S}$$
(6)

where *t* is the time and x_i the spatial coordinate. The index i = 1, 2, 3 representing the spatial dimension is summed over. The term \dot{Q}_S on the right hand side of equation (6) represents the energy source terms due to heat exchange of the solid fraction of the packed bed with the fluid (see section 3.5). The porosity and the particle density in each cell determine the bulk density

$$\rho_{\rm S} = (1 - \varepsilon) \cdot \rho_{\rm P} \tag{7}$$

of the packed bed. For describing the effective thermal conductivity λ_S of the packed bed, the model proposed in Ref. [9] is used, which is based on the heat-electrical analogy, and the concept of Hertzian contact between spheres:

$$\lambda_{s} = G_{s} \frac{l_{e}}{d_{p}} \tag{8}$$

$$G_S = \frac{G_P G_C}{G_P + G_C} \tag{9}$$

$$G_P = \frac{1}{4} \lambda_P l_s \pi \, d_p \tag{10}$$

$$G_C = 2\lambda_P \left(\frac{3F_n d_p}{2E^*}\right)^{\frac{1}{3}} \tag{11}$$

here, G_S, G_C and G_P are the conductance of the packed bed, at contact points between particles and of the particles, respectively, with $l_s = 2.0$, $l_e = 0.9$ from Ref. [11] being geometric factors, $E^* = 11.761$ GPa is the effective Young's modulus from Ref. [12] and F_n is an estimate of the normal forces between the (spherical) particles of the fuel bed. It is calculated from the number of contact points between the particles and the pressure exerted from the weight of the fuel bed: The number of particles in the retort can easily be obtained from the total volume of the packed bed, the porosity, and the particle diameter:

$$n_p = \frac{V_{tot}(1-\varepsilon)}{V_p} \tag{12}$$

It is then assumed, that the particles are assembled in layers according to a dense spherical packing. Under this circumstance, the number of particles in one layer can be estimated as

$$n_{layer} = n_p \frac{d_p}{h_{bed}} \tag{13}$$

from the particle diameter d_p and the total height of the fuel bed h_{bed} . In the dense packing, each sphere is in direct contact with three spheres from the layer above. For the whole layer, the number of contact points is therefore given by $n_{contact} = 3 \cdot n_{layer}$. The pressure *P* caused by the mass of the fuel bed above a given location is given by the bulk density of the

Table 1

Ultimate analysis (mass fractions) of the raw material (beech wood chips) used for the selected test run (mass fractions of C, H, O, and N are given on dry basis, the water mass fraction on wet basis)..

Element	Value	Unit
С	48.7	%
Н	6.0	%
0	44.8	%
Ν	0.4	m%
Water	10.8	%

bed, ρ_{bed} , and the distance from the specific location to the surface of the fuel bed, h_{local} , as

$$P = \rho_{bed} h_{local} \cdot g \tag{14}$$

 $(g = 9.81 \text{ m/s}^2$ is the gravitational acceleration). The normal forces are then given by the pressure distributed over the contact points over the cross section of the fuel bed, A_{CS} :

$$F_n = \frac{PA_{CS}}{n_{contact}} \frac{\sqrt{2}}{2} \tag{15}$$

3.3. Drying and pyrolysis

At the start of the process, the fuel particles consist of moisture and dry wood, according to the water mass fraction of the biomass used as fuel. During drying, the moisture is released to the gas phase:

$$H2O \rightarrow H2O(g)$$
 (16)

If the temperature of the packed bed is above 100 °C, and energy is transferred to the solid, the energy does not heat up the solid, but is consumed by the evaporation of moisture from the particle (if present). The volumetric source term describing the amount of water released from the particle is calculated from the energy ΔH transferred to the particle via

$$\dot{m}_{H2O} = \frac{\Delta H}{H_{dry}} \tag{17}$$

where $H_{dry} = 2.26 \cdot 10^6$ J/kg is the latent heat of evaporation of water at 100 °C.

Below a temperature of 100 °C, the drying rate is calculated according to a "drying level" model [13], that was adapted and validated by BIOS. The drying rate depends on the gas phase saturation properties, the water mass fraction of the particle, as well as the transport coefficients of water in the gas phase. Depending on the water content of the biomass, three stages of drying are considered: In the first stage, the water mass fraction is higher than the fibre saturation point. In this situation, the water is easily transported to the surface of the particle, leading to a fast evaporation. In the second stage, the drying rate is reduced, while the surface of the particle continues to dry and an equilibrium between the surface water content and the surroundings is established. In this stage of drying, the drying level moves from the surface of the particle gradually towards the centre. The third stage starts as soon as the drying level has reached the centre of the particle. Afterwards, the drying continues more slowly, as the water mass fraction continues to decrease. Condensation of water on the biomass particles is not considered.

In the present work, the pyrolysis process is described according to a three-component model based on experimental TGA analyses of the respective fuel (see also [14]). The pyrolysis process can start in one computational cell as soon as the water is evaporated. The dry wood (DW) present in the particle is then instantly converted to the so-called pseudocomponents of Hemicellulose (HC), Cellulose (CE), and Lignin (LI):

$$DW \rightarrow CE + HC + LI$$

(18)

Table 2

Initial conditions for the simulation. The gas composition of air is given in mass fraction on wet basis.

Value	Unit
8	°C
0.553	-
10.8	%
3.5	cm
23.15	%
0.05	%
76.8	%
	Value 8 0.553 10.8 3.5 23.15 0.05 76.8

Table 3

Kinetic parameters and fractions of the pseudocomponents (Hemicellulose, Cellulose, Lignin) from a TGA analysis of beech wood chips. (E_a : activation energy, k_0 : pre-exponential factor, a.f.: ash free).

Component	Mass fraction (on dry basis, ash free)	Ea	ln(k ₀)
Units	%	kJ/mol	s^{-1}
Hemicellulose	26.4	128	22.9
Lignin	20.1	204.8 47	35.6 3.1

The relative amounts of the pseudocomponents are given by the TGA analysis (cf. section 4.3, Table 3). Hemicellulose, Cellulose, and Lignin behave like dry wood (they have the same thermal conductivity, specific heat, and density), and the contribution of these components to the particle volume is calculated via equation (1). The total particle volume at this stage of the conversion process is given by $V_P = V_{CE} + V_{HC} + V_{LI} + V_{CH}$.

These pseudocomponents are converted to volatiles VOL(g) which are released into the gas phase, and charcoal (*CH*) which remains in the solid phase:

$$CE \rightarrow (1 - C_{res}) VOL(g) + C_{res} CH$$
 (19)

$$HC \rightarrow (1 - C_{res}) VOL(g) + C_{res} CH$$
 (20)

 $LI \rightarrow (1 - C_{res}) VOL(g) + C_{res} CH$ (21)

(these equations refer to the masses of the respective components, and C_{res} is the fraction of charcoal that is produced from the pseudocomponents, also referred to as "residual charcoal"). The conversion rates are determined by the results of the TGA analysis, and the decrease of mass of component *i*, $\dot{m}_{i,P}$, in the particle in the time span Δt is given by

Table 4Composition of the volatiles released during pyroly-sis (measurements from the representative test runselected as a basis for the simulation). Values aregiven in mass fractions on wet basis.

Species	Value
CO ₂	16.37 %
H ₂ O	28.10 %
CO	8.43 %
CH ₄	2.23 %
H ₂	0.05 %
Ethylene	0.43 %
Gravimetric tars	27.63 %
Formaldehyde	0.24 %
Propylene	0.38 %
Acetic acid	9.87 %
Acetaldehyde	0.75 %
Methanol	4.00 %
Ethanol	0.83 %
Acetone	0.68 %

$$\dot{m}_{i,P} = m_i \cdot \frac{\left(-1 + \exp\left(-\Delta t k_{0,i} e^{-\frac{E_{a,i}}{RT_S}}\right)\right)}{\Delta t}$$
(22)

where m_i is the current mass of component i in the representative particle, $k_{0,i}$ is the pre-exponential factor, $E_{a,i}$ the activation energy, T_S the packed bed temperature, and R the universal gas constant. This equation represents a first order kinetic equation which has been integrated over the timestep Δt . This form of the equation was chosen for the model for reasons of numerical stability. The mass of the particle during the pyrolysis (assuming that all water is evaporated) is therefore given by

$$m_P = m_{CE} + m_{HC} + m_{LI} + m_{CH} \tag{23}$$

The volatiles VOL(g) are released into the gas phase as a mixture of chemical species. The composition is kept constant (see also Table 4). Also note that no reaction enthalpy for pyrolysis is considered in the present model.

3.4. Radiation

To describe the interaction of the radiation with the packed bed as well as with the gas phase, the radiative transfer equation is modified. This approach, detailed in Ref. [9], has also been used in Ref. [15] in a CFD model to describe a porous burner.

The total absorption coefficient is given by a contribution from the packed bed, α_s , and a contribution from the gas α_g :

$$\alpha = \alpha_g + \alpha_s \tag{24}$$

The absorption coefficient of the packed bed is modelled as

$$\alpha_{S} = \varepsilon_{P} \left(-\frac{1}{d_{p}} \ln(\varepsilon) \right)$$
(25)

where ε_P is the emissivity of the particle surface, d_p the particle diameter, and ε the packed bed porosity. Along the lines of [9], scattering of radiation in the packed bed is considered as well, with a scattering coefficient of

$$\sigma_{scat} = (1 - \varepsilon_P) \left(-\frac{1}{d_p} \ln(\varepsilon) \right)$$
(26)

Scattering in the gas phase is neglected.

3.5. Heat transfer between solid and gas phase

In the transport equation for the solid phase temperature, eq. (6), the source term on the right hand side describes the heat transfer between the particles and the gas phase via convection and mass transfer (represented by the symbols $\dot{Q}_{S,conv}$ and $\dot{Q}_{S,mdot}$, respectively) as well as the radiative contribution to the heat transfer, $\dot{Q}_{S,rad}$:

$$\dot{Q}_{s} = \dot{Q}_{s,conv} + \dot{Q}_{s,rad} + \dot{Q}_{s,mdot}$$
⁽²⁷⁾

The convective source term is given by

$$\dot{Q}_{s,conv} = Ah(T - T_s) \tag{28}$$

with the surface area per volume

$$A = \frac{\pi d_p^2}{V_P} (1 - \varepsilon)$$
⁽²⁹⁾

and the heat transfer coefficient

$$h = N u \frac{\lambda_g}{d_p} \tag{30}$$

which is determined by the thermal conductivity of the gas λ_g , and the

Nusselt number Nu.

A correct approach for the Nusselt number is crucial for modelling the convective heat transfer. There is no flow of air through the packed bed inside the pyrolysis reactor, only the gas released from the particles has to be considered. Thus, very low flow velocities of the order of 0.1 m/s and less can be expected (indeed, numerical evaluations show a maximum velocity of 0.5 m/s inside the retort). In this flow regime of low Reynolds and Peclet numbers, the Nusselt number is significantly smaller than the one obtained from correlations derived from flows with higher Reynolds number. In Refs. [16], this has been explained by differences in the packing density (and thus the porosity), which have significant influence on the flow velocities, if the Peclet number is low. Based on the Ergun equation for the pressure loss in packed beds, and the assumption that a packed bed shows areas of higher and lower packing density, Martin derived an equation for the Nusselt number, valid for low and high Peclet-number flows (we refer the reader to Ref. [16] for a detailed explanation and derivation of the equation):

$$Nu = \frac{1}{Nu_1} + \varphi^* \left(\frac{1}{Nu_2} - \frac{1}{Nu_1} \right) + \frac{AL}{Pe} \frac{\nu}{1 - \nu} \left(1 - \frac{\varphi^*}{\nu} \right)^2$$
(31)

Here, Nu_1 and Nu_2 are the Nusselt numbers in the parts of lower and higher porosity, respectively, and

$$\varphi^* = \varphi \frac{1 - \varepsilon_2}{1 - \varepsilon}.$$
(32)

 φ is the fraction of the packed bed volume that has the higher porosity ε_2 and ν is the fraction of the volume flow that passes through the part of the packed bed with porosity ε_2 . Following [16], $\varepsilon_2 = \varepsilon + 0.1$ and $\nu = 0.06$ were used here. Further variables are the length of the packed bed *L*, the Peclet number, Pe, (the product of the Reynolds number, Re, and the Prandtl number, Pr):

$$Pe = Re \cdot Pr \tag{33}$$

$$Re = \frac{\rho_g U_0 d_p}{\mu_e \varepsilon} \tag{34}$$

$$Pr = \frac{\mu_g c_{P,g}}{\lambda_g} \tag{35}$$

as well as the superficial velocity U_0 , and the gas properties ρ_g , μ_g , $c_{p,g}$, λ_g (density, viscosity, heat capacity, thermal conductivity), the surface area of the packed bed particles divided by the total volume *A*, and the overall porosity ε , which is calculated according to the model via equation (4).

 Nu_1 and Nu_2 are calculated separately for their corresponding porosities, via the correlation given in Ref. [17] for packed beds of spherical particles:

$$Nu = (1 + 1.5(1 - \varepsilon)) \left(2 + \sqrt{Nu_{lam}^2 + Nu_{larb}^2} \right)$$
(36)

$$Nu_{lam} = 0.664 \sqrt{Re} + Pr^{\frac{1}{3}}$$
(37)

$$Nu_{turb} = 0.037 \frac{Re^{0.8} Pr}{1.0 + 2.443 Re^{-0.1} \left(Pr_{5}^{2} - 1.0 \right)}$$
(38)

The packed bed temperature interacts with the radiative transfer equation via an energy source term [9]

$$\dot{Q}_{S,rad} = \alpha_S G_{rad} - 4\alpha_S \sigma T_S^4 \tag{39}$$

allowing for radiative heat transfer to and from the packed bed (note that this equation is only strictly valid if the variables in the second term are constant). Here, G_{rad} is the total incident radiation, α_S the solid absorption coefficient described in section 3.4, and σ the Stefan-Boltzmann constant.



Fig. 3. Positions of the measurement points M1 - M6 for temperature measurements inside the retort (all units given in mm).

The last source term corresponds to the heat transfer via mass exchange between the solid and the gas phase, as well as contributions from the heat of evaporation. It is given by

$$\dot{Q}_{S,mdot} = \dot{Q}_{S,Pyro} + \dot{Q}_{S,Dry},\tag{40}$$

$$\dot{Q}_{S,Pyro} = \dot{m}_{CH} \int_{T_{ref}}^{T_S} c_p^{CH}(T) dT + \dot{m}_{VOL} \int_{T_{ref}}^{T_S} c_p^{DW}(T) dT,$$
(41)

$$\dot{Q}_{S,Dry} = \dot{m}_{H2O} \int_{T_{ref}}^{T_{H2O}} c_p^{H2O}(T) dT + \dot{m}_{H2O} H_{dry}.$$
(42)

The species are released to the gas phase at the packed bed temperature T_s , except the water from drying, which is released at T_{H2O} =100 °C.

3.6. Numerical aspects

The advantage of the model presented here over a particle-based

approach lies in its comparative simplicity. The model equations can easily be integrated in the framework of the CFD software in the form of scalar transport equations. In total, there are eight additional transport equations, which have to be solved: equation (6) for the packed bed temperature, equation (4) for the porosity, as well as equations for the masses of water, dry wood, the pseudocomponents (hemicellulose, cellulose, lignin), and charcoal in the representative particles. This formulation allows numerically reliable and stable calculations, with a moderate time-step size of 10 s, and no model-specific restrictions on the size of the computational cells.

In contrast, for the particle-based models, the equation of motion for each particle needs to be solved to obtain a description of particle movement. In addition, for each particle, the drying and pyrolysis processes has to be calculated for the whole trajectory. Especially in the context of a packed bed, the paths of the particles are not easily calculated: The movement is only caused by the shrinkage of the fuel bed, and thus the individual velocities of the particles are small. Therefore, to properly resolve this movement, a high resolution is needed, leading to very small time-steps (of the order of 0.001 s) and computational cells, such that the computational time needed to simulate large-scale processes soon becomes prohibitive.

4. Experimental and computational setup

4.1. Measurements performed

Special attention was payed to detailed continuous measurements of temperatures, both at inlet and exit pipes of the flue gas, at the exit of the pyrolysis gas, and inside the retort. There, temperature sensors were placed in six locations (cf. Fig. 3). The sensors were Type K Thermo-couples, but for the measurement positions in the pipes the thermo-couple measurements were crosschecked with suction pyrometer measurements.

The volume flow of the pyrolysis gas and the flue gas was obtained using Prandtl-tube measurements; the mass flow was then calculated with the corresponding gas density. The cross-section of the pipe as well as the correction factor of the flow profile have been considered as the gas velocity measured by the Pitot tube in the middle of the pipe is the maximum value in the pipe. The pressure difference in the Pitot tube has been measured online by a pressure transmitter of Kalinsky (DMU2). The density of the gas has been calculated from the real-time measured gas composition.

The composition of the flue gas was measured by using standard flue gas analysers of Rosemount (Emerson) for O_2 (paramagnetic sensor), CO and NO (NDIR) as well as by FT-IR (Gasmet, DX4000) for H₂O, CO, CO₂, CH₄, ethene, formaldehyde, propene, acetic acid, acetaldehyde, methanol, ethanol and acetone. Gas collection tubes (samples have been analysed by GC-MS) were used for selected species (N₂, H₂, CO, CO₂, O₂, CH₄, ethene) to double-check the continuous data, and verify the calculated concentrations of N₂. Data for H₂ was obtained solely using gas collection tubes measurements. The tar mass fraction of the pyrolysis gas was obtained using gravimetric methods ("tar protocol", CEN TC BT/TF 143 WICSC 03002.4; 2005); several tar measurements over the whole batch have been performed. Before and after the batch, the mass of the contents of the retort was determined, as well as the corresponding bed heights.

In order to assess the progress of the conversion inside the retort during the batch, three interruption tests were carried out. For each test, the batch was started as normal, then after a certain period of time (80 min, 110 min, and 130 min after the beginning of the batch) the retort was removed from the reactor. Immediately, water-cooling was applied to the outer walls of the retort. After cooling, the retort was opened, and the packed bed removed layer by layer. This was documented visually (photographs of the layer surfaces, see Fig. 9), and samples were taken for each layer in the middle of the retort, as well as near the walls and at an intermediate position for future analysis.

4.2. Simulation domain

The batch reactor was simulated as depicted in Fig. 2. The simulation domain encompasses the whole retort with the packed bed, the pyrolysis gas channel outside the retort, and the exit pipe for the pyrolysis gas, as well as the flue gas channel outside the pyrolysis gas channel, with inlet and exit pipes. The insulation layers were also considered explicitly.

The cover of the retort is not completely airtight, such that a small amount of false air enters the retort through a small gap between the retort and the cover (see also Fig. 2). From the test run balances, a mass flow of false air of approximately 0.79 kg/h can be deduced. It enters the domain in a diffuse manner (as a volumetric source term) in the volume above the cover of the retort. It can enter the regions filled with pyrolysis gas via a small gap between the retort cover and the body of the retort.

In the test run, the retort was completely filled with biomass at the beginning of the batch. During the batch, due to overall bed shrinkage, the height of the packed bed decreases, such that at the end of the batch, the resulting charcoal fills 53 % of the volume of the retort, as determined by measurements of the bed height before and after the batch. This shrinkage is not a part of the CFD model (cf. also section 3.2). Therefore, an intermediate height of the packed bed is used as an approximation. Consequently, the initial porosity of the bed is adjusted from the measured value of 0.66 to 0.553, such that the total input mass corresponds to the measured initial mass.

The system was simulated with a time step size of 10 s, and a total of 2.9 million computational cells, 201,598 of which lie in the packed bed; the computational grid was unstructured. The simulation time amounted to approximately 72 h on 12 compute nodes.

4.3. Simulation conditions and model parameters

Test runs were performed with the small-scale reactor described in section 2 in order to gain data for the validation of the CFD model. One representative test run was then simulated; the simulation results are compared to the experimental findings in section 5. For the selected test run, wood chips of beech wood with a water mass fraction of 10.8 % were used as raw material. The material was not produced on-site and used as received. No specific drying procedure was employed. The fuel ultimate analysis is shown in Table 1. The average diameter of the wood chips (determined from a representative sample) is 3.5 cm. The initial mass of the wood is 75.8 kg. From the volume of the retort and the initial mass, together with the density of the biomass, the porosity of the packed bed is calculated as 0.66.

The test reactor is heated with flue gas from a gas burner (fuel: natural gas), which is supplied at a constant temperature of 875 $^{\circ}$ C, and a mass flow of 112 kg/h.

Inside the retort, constant initial conditions were set for the temperatures, the gas composition, as well as the porosity, the biomass water mass fraction, and the particle diameter. These values were selected according to the conditions of the test run and are summarized in Table 2.

It is assumed that the fixed part of the reactor (insulation and flue gas channel) is in thermal equilibrium with the flue gas flow at the beginning of the test run. Therefore, the initial temperatures in the flue gas channel and the insulation layers, as well as the initial flow field were obtained from a steady-state simulation of the flue gas flow.

As described in section 3.3, the mass release during pyrolysis of the biomass is described according to TGA data. The TGA experiment was performed with 50 mg of dried beech wood, which was heated with a rate of 20 K/min. For the recorded mass loss data, a three-component fit was performed, assuming first-order kinetic decomposition of the pseudocomponents Hemicellulose, Cellulose, and Lignin. Only one TGA experiment was used for the analysis. The values obtained for the kinetic parameters used for this work are summarized in Table 3.

The composition of the volatile gas released during pyrolysis (see sec. 3.3) was obtained from the test run results. From the measured gas

Table 5

Properties (heat capacity c_p in J/(kg K), thermal conductivity λ in W/(m K), density ρ in kg/m³, depending on the absolute temperature T in K) of the biomass particle components dry wood (DW), charcoal (CH) and moisture (H₂O).

Property	Value or Formula	Reference
c _p		
H ₂ O	4184	
DW	1500 + T	[18]
CH	$420 + 2.09 \text{ T}$ - $6.85 \ 10^{-4} \text{ T}^2$	[18]
λ		
H ₂ O	0.597	
DW	$0.056 + 2.6 \ 10^{-4} \ \mathrm{T}$	[19]
CH	0.07	[19]
ρ		
H ₂ O	998.2	
DW	682	[20]
CH	300	[BIOS database]



Fig. 4. Comparison of the simulated profile of mass flow of the pyrolysis gas with the measured curve. The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation.

composition, the water released from drying as well as the false air entering the reactor were subtracted. The values used here are presented in Table 4.

The mass fraction of residual charcoal C_{res} on a dry basis was 32 %. This value was obtained from the residual mass of charcoal in the test run chosen as a basis for the simulation. The physical properties (heat capacity, thermal conductivity, density) of the particle components are presented in Table 5

The realizable k- ε model is used to describe the turbulent flow in the flue gas channel as well as in the pyrolysis gas channel. At the flue gas inlet pipe, a turbulent intensity of 5 % was set as a boundary condition. The mixing length is calculated from the hydraulic diameter $d_H = 0.15$ m of the pipe, according to the standard approach used by ANSYS Fluent. Inside the retort, however, the flow is laminar, and no turbulent mixing is simulated. In the whole simulation domain, no gas phase reactions are considered.

5. Simulation results and comparison to experiment

5.1. Mass release and batch duration

One important criterion for evaluating the results of the CFD simulation of the batch reactor is the characteristics of the mass release from the packed bed, i.e. the mass flow of the pyrolysis gas, shown in Fig. 4. In

Table 6

Mass balance in kg (CFD simulation compared to experiment).

	CFD simulation	Experiment
Mass input		
Biomass	75.9	75.8
False air	1.3	3.0
Total	77.1	78.8
Mass output		
Pyrolysis gas	54.9	55.0
Charcoal	22.1	23.4
Total	77.0	78.4



Fig. 5. Comparison of simulated and measured temperatures of flue gas (solid lines) and of pyrolysis gas at the exit of the reactor (dashed lines). The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation. Remark: The fluctuations of the measured flue gas temperature at a batch duration of approx. 1:45 are due to fluctuations of the inlet temperature of the flue gas (operational fluctuations).

the first 45 min of the batch, the simulated mass flow agrees well with the measured values. After that, the simulation shows higher mass release than the experiment, with a peak mass flow of 63 kg/h for the simulation, compared to 35 kg/h according to the measurement. Consequently, the simulated batch duration of 2 h 15 min (defined at the point where the measured mass flow of the pyrolysis gas at the end of the batch agrees with the simulated one) is shorter than the measured 3 h. The mass balances of the simulated and the measured system are given in Table 6, and show a quite good agreement. Furthermore, the closure of the total mass input) is a very important indicator of the excellent numerical quality achieved.

5.2. Temperatures and energy balance

A comparison of simulated and measured values of flue gas and pyrolysis gas temperatures at the exit of the reactor is shown in Fig. 5. The simulated flue gas temperature agrees quite well with the experimentally determined value, showing that the overall heat transfer into the retort is simulated correctly. The temperature of the pyrolysis gas



Fig. 6. Comparison of measured and simulated temperatures at sensor M2 (cf. Fig. 3). Note that solid and gas phase temperatures cannot be measured separately. The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation.



Fig. 7. Comparison of measured and simulated temperatures at sensor M5 (cf. Fig. 3). Note that solid and gas phase temperatures cannot be measured separately. The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation.

shows larger deviations between simulation and experiment, which are mainly due to the differences in mass flow, as explained in section 5.1. This results in a cooling effect on the outer wall of the retort (inner wall of the pyrolysis gas channel), thereby reducing the amount of heat transferred to the material inside the retort.

Inside the retort, near the walls, the measured temperatures at the sensors M1 and M2 (positions: see Fig. 3) are in very good agreement with the simulated gas phase and solid phase temperatures (note that the

Table 7

Energy input and output for the batch in MJ (CFD simulation compared to experiment).

	CFD simulation	Experiment
Energy input via flue gas	124.5	144.7
Energy output		
Sensible enthalpy of pyrolysis gas	41.3	31.3
Sensible enthalpy of the charcoal and the retort (end of batch)	40.1	47.9
Heat losses to environment	32.8	41.0

solid and the gas phase temperatures can in practice not be measured separately). The comparison is shown in Fig. 6 for the sensor M2. All curves show a clear plateau at 100 °C due to drying.

Near the centre of the retort (sensors M4 and M5, see Fig. 7 for M5), larger deviations are visible. The measured temperature curve shows a steep increase around 30 min after the start of the batch, followed by a very long plateau, until the temperature continues to increase around 2 h after the beginning of the batch. The simulated curves have a much smaller slope in the first phase of the batch, and the plateau is significantly shorter. After the plateau, the simulation appears to be shifted with respect to the experiment by approximately 30 min, corresponding to the shorter batch duration.

This behaviour can be interpreted as an effect of condensation. Early in the batch, the water evaporates in the hotter regions of the packed bed, near the walls. The water is then transported through the bed towards the exit of the retort located in the centre at the bottom. There, the temperatures are significantly lower, and the saturated gas condenses. The enthalpy released in this process causes an increase of the temperatures, until an equilibrium is reached. Thus, the temporary water mass fraction in the centre of the retort is higher than near the walls, and the pyrolysis process is slowed down in this area. This is consistent with the observation that the simulated batch duration is shorter than the measured batch duration. It is planned, however, to extend the model to include condensation, such that these aspects can be better described in the future. At later stages of the model improvement, also the effect of a non-zero heat of pyrolysis may be studied.

A comparison between the simulation and the experiment for the energy input and output over the batch are shown in Table 7. The measured value of energy transferred to the retort by the hot flue gas is



Fig. 8. Comparison of measured and simulated profiles of H_2O concentrations in the pyrolysis gas at the exit of the reactor over the batch. The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation.



Fig. 9. Comparison of measured and simulated profiles of CO_2 concentrations in the pyrolysis gas at the exit of the reactor over the batch. The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation.



Fig. 10. Comparison of measured and simulated profiles of CO concentrations in the pyrolysis gas at the exit of the reactor over the batch. The dashed line at 02:15 [hh:mm] indicates the end of the batch according to the simulation.

20.2 MJ larger than the simulation result, which can be attributed to the shorter batch duration in the simulation. Because of this, the exposure time of the whole system to the hot flue gas flow is smaller in the simulation, and thus less energy is transferred. In contrast, the sensible enthalpy leaving the reactor via the hot pyrolysis gas is larger (by 10 MJ) in the simulation than in the experiment. This is probably caused by the enhanced flow of the pyrolysis gas, which leads to a more efficient convective heat transfer and thus to a larger cooling effect. This is consistent with the results for the sensible enthalpy of the charcoal and retort at the end of the batch, which is smaller in the simulation. The differences in the heat losses to the environment are also due to the differences in the batch duration.

30 cm from the surface of the packed bed:



50 cm from the surface of the packed bed:



70 cm from the surface of the packed bed:



Fig. 11. Photographs of the packed bed resulting from the batch interruption test after 80 min (left) compared to CFD simulation results regarding the char mass fraction inside the retort [m% d.b.] (right) at approximately the same time. Top: 300 mm below the original bed surface; Middle: 500 mm below the original bed surface; Bottom: 700 mm below the original bed surface. The white dash-dotted lines in the photographs highlight the walls of the retort. All pictures (photographs and CFD results) are at the same scale.

5.3. Species concentrations

The effects of condensation are also apparent when comparing the profile of the concentration of H_2O in the pyrolysis gas, as shown in Fig. 8. At the beginning of the batch, the simulation exhibits larger concentrations of H_2O , indicating that a significant amount of water, which is evaporated in the outer (near-wall) parts of the packed bed, does not leave the reactor, but condenses in the inner (colder) parts of the packed bed. After approximately 30 min, the simulated and measured water mass fractions of the gas agree. Later on (after 1 h), the simulated curve lies below the experimental one, indicating that the previously condensed water is released during that part of the batch.

At the beginning of the batch, the experimental curves for the concentrations of CO and CO2 show large peaks, which the simulation does not reproduce, as shown in Fig. 9 for the case of CO₂ and in Fig. 10 for CO. This effect can be attributed to gas phase combustion reactions, which are most prominent in the starting phase of the batch, because the air filling the retort at the beginning is not yet replaced by pyrolysis gas. The increase of the CO and CO₂ concentrations in the pyrolysis gas observed in the simulation corresponds to the onset of the pyrolysis in a significant portion of the raw biomass approximately 30 min after the beginning of the batch. Before this, the pyrolysis gas is dominated by the water released due to drying. The peaks of CO and CO2 in the experimental curves, however, appear much earlier (approximately 10 min after the beginning of the batch). This indicates that the start of the pyrolysis reactions in the simulation is delayed. A likely reason for this is the missing combustion reactions caused by a certain amount of false air intake in the testing plant, which lead to increased temperatures and faster pyrolysis at the beginning of the batch. These reactions will be included in the next step of model development.

In later parts of the batch, the CO_2 and CO concentrations are at a similar level and show a similar overall trend with the progress of the batch.

5.4. Degree of conversion

The progress of the pyrolysis inside the packed bed shows an excellent agreement between the CFD simulation and the batch interruption tests. The pyrolysis starts near the hot walls and the conversion progresses inwards, as well as downwards from the surface of the packed bed. As exemplary result, a direct comparison of the photographs taken after the batch interruption test after 80 min with CFD results for the charcoal mass fraction in the packed bed is shown in Fig. 11. The

Nomenclature table

Latin letters

A	surface area per volume (1/m)
CE	cellulose
CH	charcoal
c_p^p , $c_{p,g}$	particle heat capacity, gas phase heat capacity (J/(kg K))
Cres	residual charcoal
dp	particle diameter (m)
DW	dry wood
E*	effective Young's modulus (Pa)
E _{a,i}	activation energy of component i (J/mol)
F _N	normal force (N)
G_S, G_C, G_I	e conductance of the packed bed, at contact points between particles, of particles (W/K)
G _{rad}	total incident radiation (W/m ²)
HC	hemicellulose
H _{dry}	latent heat of evaporation of water at 100 °C (J/kg)
h	heat transfer coefficient (W/m·K)
k ₀	first order pre-exponential factor (1/s)
LI	lignin

simulation as well as the photographs show a more advanced pyrolysis on the left side, which corresponds to the hotter parts of the retort on the side opposite the inlet pipe of the flue gas which is due to the flow pattern of the hot flue gas around the pyrolysis gas channel. The similarity prevails, even though in the simulation the shrinkage of the bed was not considered, and a constant height of the fuel bed of 0.73 m was assumed throughout the batch.

6. Summary

The newly developed CFD model for the description of the pyrolysis in a batch reactor for charcoal production exhibits reasonable agreement with experimental data obtained during dedicated test runs. The local progress of the pyrolysis (downwards from the surface of the packed bed, and inwards from the hot walls) is reproduced very well. The data for temperatures in the near-wall region of the packed bed are also in reasonable agreement with the experimental results.

The present results also clearly highlight the directions to be taken for further improvements of the CFD model. The next steps will be the consideration of condensation, which is expected to increase the batch duration, as well as the gas phase reactions which are of importance to correctly describe the measured species concentrations in the pyrolysis gas. At later stages of the model improvement, also the effect of a nonzero heat of pyrolysis may be studied.

In addition, the model is computationally efficient, and thus allows detailed analyses of large-scale applications.

7. Conclusion

The CFD model presented in this work has the capability to simulate the production of charcoal in a batch reactor of industrial scale. It was successfully applied during the project to optimize the pyrolysis reactor, and studies were performed concerning the effects of geometric variations on the conversion process. Thus, the model provides a valuable tool, and could contribute to reach the project goal regarding the development of a new and highly efficient technology for the large-scale production of high-quality charcoal.

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- L length of packed bed (m)
- l_s, l_e geometric parameter (–)
- m_i mass of component i (kg)
- m mass source term(kg/($m^3 \cdot s$))
- Nu, Nu_{lam}, Nu_{turb} Nusselt number (total, laminar, turbulent) (–)
- Pe Peclet number (–)
- Pr Prandtl number (-)

Q's, Q's, conv, Q's, rad energy source terms (total, convection, radiation, mass transfer) (W)

- Q_{S,mdot}, Q_{S,Pyro}, Q_{S,Dry} energy source terms due to mass transfer (total, pyrolysis, drying) (W)
- R universal gas constant (J/(mol·K))
- Re Reynolds number (–)
- T_S, T_{ref} packed bed temperature, reference temperature (K)
- t time (s)
- U₀ superficial gas velocity (m/s)
- V_p, V_i, V_{DW}, V_{CH} Volume of particle, component i, dry wood, charcoal (m³)
- VOL volatiles
- x_{1,2,3} spatial coordinates (m)

Greek letters

- α , α_g , α_S absorption coefficient (total, gas phase, packed bed) (1/m)
- Δp pressure loss of packed bed (Pa)
- ΔH heat (W)
- ε fluid porosity of the packed bed (m³/m³)
- $\epsilon_{\rm P}$ particle emissivity (-)
- $\lambda p, \lambda_S$ particle thermal conductivity, packed bed thermal conductivity (W/(m·K))
- μ_g gas phase dynamic viscosity (Pa·s)
- ν fraction of volume flow through packed bed with higher porosity
- ρ_p , ρ_i , ρ_g particle density, density of component i, gas phase density (kg/m³)
- σ_{scat} scattering coefficient (-)
- σ Stefan-Boltzmann constant (W/(m²·K⁴))
- φ fraction of packed bed volume with higher porosity

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