Simulation of biomass particle evolution under pyrolysis conditions using lattice Boltzmann method

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Abstract

To accurately characterize biomass fast pyrolysis at the particle scale, intra-particle transport phenomena needs to be considered together with the gas flow surrounding the particle. In this study, a detailed numerical method was used to simulate the evolution of biomass particles under fast pyrolysis conditions. To conduct particle-scale simulations, the lattice Boltzmann method (LBM) was employed to solve the conservation equations, and pyrolysis kinetics were implemented to describe the chemical reactions. The present model was validated by comparing the numerical results with experimental data for a single biomass particle under pyrolysis conditions. The predicted temperature and conversion history agree with the experimental data. The temperature and density fields in the particle were found to be anisotropic due to the effect of the gas flow surrounding the particle, which was ignored by the 1D model. The non-uniform distributions of surface temperature indicate that using a constant temperature or heat flux as boundary conditions may cause numerical errors. Sensitivity analysis shows that density is the most influential parameter while porosity is the least. The heat of reaction converting the intermediate solid to char is more dominant than those of the three primary reactions. A parametric study was conducted to investigate the effect of particle shape on conversion time and final product yields. The simulation results show that the conversion time decreased when using the elliptic particle instead of the regular (circular) particle. The model

also shows that more tar and syngas were produced, while less char was generated from an elliptic particle. The effect of particle shape on the center temperature of the biomass particle can be explained by comparing the heat transfer conditions at the front of the particle and the convective heat transfer at the top and bottom of the particle. The results demonstrated that the current LBM framework has the ability to reveal the detailed conversion process of biomass particles under various pyrolysis conditions, which can then be used to improve engineering models for reactor-scale simulation.

Keywords: Biomass fast pyrolysis; Lattice Boltzmann method; Particle scale modeling

1 Introduction

Biomass is regarded as a promising alternative energy source to traditional fossil fuels because of its renewable nature and abundant supply [1]. To obtain high-energy density products, thermochemical conversion of biomass is often utilized to convert raw biomass materials into liquid biofuels (which can be further refined to produce transportation fuels) and other hydrocarbon gases [2]. As a prominent thermochemical conversion strategy, fast pyrolysis has attracted significant attention [3, 4]. Generally, in the absence of oxygen, lignocellulosic biomass particles are delivered into a reactor, and rapidly heated and decomposed in a very short time into three major products: condensable gases (tar), char, and light gases (syngas), making fast pyrolysis a feasible option for various applications.

Biomass fast pyrolysis in a reactor is a complex process, in which multiphase hydrodynamics is coupled with multiple chemical reactions. To investigate the characteristics of this process, many experimental [5-8] and computational [9-17] studies have been conducted.

Numerical simulation studies have proven to be an effective and economical alternative to time consuming and expensive experimental studies. Computational fluid dynamics (CFD) has been widely used for studying the mechanisms of biomass pyrolysis and optimizing reactor designs and operation conditions [18-24]. For reactor-scale CFD modeling, engineering models with primarily empirical coefficients are often required [22, 23]. Inability to reveal intra-particle evolution can sometimes cause numerical errors [10]. Because of the multi-scale nature of biomass feedstock and the complexity of the multiphase flow in the reactor, fundamental mechanisms of biomass pyrolysis still remain unknown [25]. Therefore, it is necessary to understand the complex processes of biomass fast pyrolysis at the particle scale, especially for large particles which are considered thermally-thick [26].

Developing a method capable of accurately predicting the details of biomass fast pyrolysis at the particle scale is challenging [25, 27]. The most popular method for simulating biomass fast pyrolysis at the particle scale is the single particle model, which is one-dimensional (1D) [12, 16, 19, 26-29]. Although the 1D model is effective in presenting the evolution process in the particle, it can suffer from several limitations. First, the boundary conditions are predefined, for instance, setting a constant temperature or heat flux on the surface, which may not be appropriate, considering the effect of gas flow surrounding the particle in a practical reactor. Besides, in 1D simulations, it is assumed that the transport of heat and mass is homogenous and isotropic in the particle. However, since the surrounding gas will significantly affect the heat transfer between gas and particles, the temperature and density fields within biomass particles will not be uniform when undergoing fast pyrolysis. In the literature, only a few studies have attempted to consider the interaction between gas and particles by coupling the intra-particle transport with the surrounding gas flow [30-32]. Another shortcoming of the 1D model is that it is intrinsically

unable to simulate particles with different shapes, such as the elliptic and ellipsoid shapes. It is important to consider the shape of the biomass particle during the fast pyrolysis process [33, 34].

The objective of this work was to assess a numerical approach to model biomass fast pyrolysis at the particle scale. This numerical approach has the potential to address the aforementioned problems and to improve our understanding of the fast pyrolysis process. The approach is based on the lattice Boltzmann method (LBM) to solve for the mass, momentum and heat transport for both gas and solid phases at the particle scale. The LBM framework was first validated by comparing the simulated results with various sets of experimental data, for a single biomass particle, under practical pyrolysis conditions. Then, a parametric study was conducted to investigate the effect of particle shape on the conversion process of a biomass particle (e.g., conversion time and final product yields).

2 Numerical methods

This section describes the lattice Boltzmann formulations used for simulating particle-scale biomass fast pyrolysis. This method offers the advantage of resolving the transport phenomena inside and outside the biomass particle altogether in an efficient manner. The Eulerian and explicit nature of LBM makes it suitable for fluid-particle problems. The pressure term in LBM is obtained by using an equation of state rather than solving the Poisson equation, as in conventional methods. This feature allows LBM to be implemented easily. LBM is also suitable for complex geometry and boundary. Additionally LBM is inherently parallel, the lattice collision is executed independently, and the propagation of the flow information only requires the relevant distribution functions from the nearest lattices.

Regarding the intra-particle transport phenomena, the following assumptions were adopted for simulating the gas flow, and the heat and mass transfer within the reactor using LBM.

- (1) The external forces (e.g. gravity) are not considered as the particle is fixed in the reactor, the surrounding gas is ideal and incompressible, and the flow is laminar.
- (2) The porous structure in the biomass particle is assumed homogenous and isotropic. The gas flow in the particle is modeled using the Darcy's law.
- (3) Particle shrinkage was not considered, since the effect of the shrinkage on the mass loss rate was not significant [26].
- (4) The thermophysical properties of the components are constant [35]. Thermal conductivity of the local solid mixture is linearly interpolated between the virgin biomass and the char formed.

$$\lambda_s = (1-\eta)\lambda_w + \eta\lambda_c \quad (1)$$

The conversion extent of pyrolysis is defined as $\eta = 1 - \frac{\rho_{ucw}}{\rho_w}$, ρ_{ucw} and ρ_w are the non-charring and virgin biomass density.

- (5) The local thermal equilibrium was assumed between the solid and gas phase, e.g., no temperature difference between the gas and solid interface in the particle [15, 26].

2.1 Governing equations

Generally, the thermochemical conversion process of biomass particles in the reactor can be described by a set of conservation equations [15, 23]. These governing equations for flow, heat, and mass transfer are as follows.

1. Conservation equations for the surrounding gas in the reactor are,

$$\frac{\partial \rho_g}{\partial t} + \nabla \cdot (\rho_g \mathbf{u}_g) = 0, \quad (2)$$

$$\frac{\partial (\rho_g \mathbf{u}_g)}{\partial t} + \nabla \cdot (\rho_g \mathbf{u}_g \mathbf{u}_g) = -\nabla p + \mu_g \nabla^2 \mathbf{u}_g, \text{ and} \quad (3)$$

$$\frac{\partial (\rho_g C_{Pg} T_g)}{\partial t} + \nabla \cdot (\rho_g C_{Pg} T_g \mathbf{u}_g) = \lambda_g \nabla^2 T_g. \quad (4)$$

2. Governing equations for intra-particle heat and mass transfer are

$$\frac{\partial \rho_s}{\partial t} = \omega_s, \quad (5)$$

$$\frac{\partial (\varepsilon \rho_f)}{\partial t} + \nabla \cdot (\rho_f \mathbf{u}_f) = \omega_f, \text{ and} \quad (6)$$

$$\frac{\partial ((\rho_s C_{Ps} + \varepsilon \rho_f C_{Pf}) T_s)}{\partial t} + \nabla \cdot (\rho_f C_{Pf} T_s \mathbf{u}_f) = \lambda_s \nabla^2 T_s + Q. \quad (7)$$

In the above equations, ε is the porosity of biomass particle, defined as $\varepsilon = 1 - \frac{\rho_s}{\rho_w} (1 - \varepsilon_w)$. ρ_s is

the total solid masses per unit volume, ε_w is the initial porosity of the biomass particle. ω_s and

ω_f are the source terms due to chemical reactions with $\omega_s = -\omega_f$ for mass conservation. More

detailed descriptions about ω_s and ω_f will be given later according to the reaction mechanisms.

Q is the heat of chemical reactions, and $Q = \sum \Delta h_j \omega_j$.

2.2 Lattice Boltzmann method

In this study, the lattice Boltzmann method (LBM) [36] was used to simulate the transport phenomena described by the above equations. Instead of directly solving the above governing equations, Eq. (2) – (7), when applying LBM, we solve the lattice Boltzmann equations (LBE).

At low Mach number, LBE can be extended to incompressible Navier-Stokes equations with second-order accuracy via the Chapman-Enskog expansion. In the LBM, the simulation domain is discretized into regular lattices, as shown in Fig. 1(a). For simulating the flow, temperature, and density fields in the reactor, the widely-used single-relaxation-time Bhatnagar-Gross-Krook formulation [37] was employed. The evolution of the density distribution function at each time step is given by

$$g_i(\mathbf{x} + \mathbf{e}_i \Delta t, t + \Delta t) - g_i(\mathbf{x}, t) = -\frac{1}{\tau_g} (g_i(\mathbf{x}, t) - g_i^{eq}(\mathbf{x}, t)), \quad (8)$$

where $\mathbf{x}$ is the position of the lattice, t is the current time, and i is the discretized direction, $\mathbf{e}_i$ is the discrete velocity in the i th direction, Δt is the time step, τ_g is the dimensionless relaxation time related to the kinematic viscosity ν , and g_i^{eq} is the equilibrium density distribution function. In this study, the D2Q9 model was adopted, in which the equilibrium density distribution function is defined as

$$g_i^{eq} = \xi_i \rho_g \left(1 + 3 \frac{\mathbf{e}_i \cdot \mathbf{u}}{c^2} + \frac{9(\mathbf{e}_i \cdot \mathbf{u})^2}{2c^4} - \frac{3u^2}{2c^2} \right), \quad i = 0 \sim 8, \quad (9)$$

where c is the lattice speed (defined as $\Delta x / \Delta t$, in which Δx is the lattice spacing), ξ_i is the weighting factor in i -th direction. Other details of LBM can be found in a previous work [36].

The evolution of the temperature distribution function has a similar form [38],

$$T_i(\mathbf{x} + \mathbf{e}_i \Delta t, t + \Delta t) - T_i(\mathbf{x}, t) = -\frac{1}{\tau_T} (T_i(\mathbf{x}, t) - T_i^{eq}(\mathbf{x}, t)) + \xi_i \frac{Q}{\rho C_p} \quad (10)$$

where τ_T is dimensionless relaxation time related to the thermal diffusivity α , which is determined by the local properties, and Q is the source term due to chemical reactions

($Q = \sum_{i=1}^N \Delta h_i \omega_i$). The equilibrium temperature distribution function is as follows [39]:

$$T_i^{eq} = \xi_i T (1 + 3 \frac{\mathbf{e}_i \cdot \mathbf{u}}{c^2}), \quad i = 0 \sim 8 \quad (11)$$

For the mass distribution of products, only fluid phase (e.g., tar and syngas) is solved by using LBM, while the solid phase (e.g., virgin biomass and char) is calculated at the fixed lattice, in which the density change of the solid phase is time dependent, similar to the single particle model [15, 26, 28]. The densities of the solid phase will be stored at the solid node while the density of the fluid phase will propagate, and diffuse, among the solid node and the fluid node, as shown in Fig. 1(b). The evolution of the density distribution function of the fluid phase is

$$f_i(\mathbf{x} + \mathbf{e}_i \Delta t, t + \Delta t) - f_i(\mathbf{x}, t) = -\frac{1}{\tau_f} (f_i(\mathbf{x}, t) - f_i^{eq}(\mathbf{x}, t)) + \xi_i \omega_f, \quad (12)$$

where τ_f is dimensionless relaxation time related to mass diffusion coefficient D , and ω_f is the source term due to chemical reactions. The equilibrium density distribution function is the same as Eq. (9).

$$f_i^{eq} = \xi_i \rho_f (1 + 3 \frac{\mathbf{e}_i \cdot \mathbf{u}}{c^2} + \frac{9(\mathbf{e}_i \cdot \mathbf{u})^2}{2c^4} - \frac{3u^2}{2c^2}), \quad i = 0 \sim 8. \quad (13)$$

Given these property distribution functions, the macro variables, such as gas density ρ_g , velocity $\mathbf{u}$, temperature T , and species density ρ_f , can be obtained by

$$\rho_g = \sum_i g_i, \quad \rho_g \mathbf{u} = \sum_i g_i \mathbf{e}_i, \quad T = \sum_i T_i, \quad \rho_f = \sum_i f_i. \quad (14)$$

The relationship between the relaxation time and the physical properties are given as follows:

$$\nu = \frac{1}{3}c^2(\tau_g - \frac{1}{2})\Delta t, \alpha = \frac{1}{3}c^2(\tau_r - \frac{1}{2})\Delta t, D = \frac{1}{3}c^2(\tau_f - \frac{1}{2})\Delta t \quad (15)$$

Boundary conditions at the inlet are the fully-developed parabolic velocity, with peak value U_{in} , and constant temperature T_{in} , as shown in Fig. 1(a). At the outlet, constant pressure is assumed. The wall temperature T_{wall} is the same as the reactor temperature, and no-slip boundary conditions are employed. The detailed treatments for addressing the unknown distribution functions on the boundary can be found in previous studies [40-42]. In this paper, to describe the chemical reactions, simplified global pyrolysis reaction mechanisms were employed. The details of the mechanisms depend on the problems studied. The reaction rates were calculated using the first-order irreversible Arrhenius expression.

3 Results and discussion

In this paper, a classical configuration was simulated, in which a stationary particle was immersed, and heated, in hot gas for fast pyrolysis. To validate the present LBM model comprehensively, two studies [14, 15] were chosen for comparison. The simulation conditions, material properties, and simulation results corresponding to each experiment will be presented.

3.1 Validation 1: Experiment by Pyle and Zaror [14]

The experiment conducted by Pyle and Zaror [14] has been adopted by many researchers [9, 17] as the benchmark to validate their single-particle models. In the experiment, pine wood cylinders with diameter of 0.6, 1.5, and 2.2 cm, and initial temperature 303 K were pyrolyzed under reactor temperatures ranging from 643 K to 780 K. Nitrogen with constant temperature (same as the reactor temperature) and velocity flowed into the reactor and took the product out. The temporal evolutions of temperatures at different radial locations inside the particle were

measured, and the conversion extent was recorded. The one step reaction in which biomass decomposes into tar and charcoal is shown below [14].

The reaction rate is $k = A \exp(-\frac{E}{RT})$, and the physical parameters are listed in Table 1. The values of heat capacity and thermal conductivity are taken from Perry and Chilton [43]. By applying the conservation equations Eq. (5)–(7), the density of each component and the temperature inside the particle can be calculated.

$$\text{Virgin wood:} \quad \frac{\partial \rho_b}{\partial t} = \omega_b = -k \rho_b \quad (17)$$

$$\text{Charcoal:} \quad \frac{\partial \rho_{ch}}{\partial t} = \omega_{ch} = k \rho_b \times \frac{\rho_c}{\rho_w} \quad (18)$$

$$\text{Tar:} \quad \frac{\partial(\varepsilon \rho_t)}{\partial t} + \nabla \cdot (\rho_t \mathbf{u}_f) = \omega_t = k \rho_b - k \rho_b \times \frac{\rho_c}{\rho_w} \quad (19)$$

$$\text{Temperature:} \quad (\rho_b C_{P_w} + \rho_{ch} C_{P_c}) \frac{\partial T_s}{\partial t} + \nabla \cdot (T_s \mathbf{u}_f) = \lambda_s \nabla^2 T_s \quad (20)$$

The experimental and numerical temperature profiles, with respect to the radial position in the particle, are shown in Fig. 2(a). The radial position was defined as the ratio of the distance from the center to the radius of the particle (e.g., 0 for center, 1 for surface). It can be seen that the simulation results agree well with the experimental data. The predicted temperatures are slightly lower than those from the experiment, which may be due to the fact that the radiative heat transfer was not considered in our model. Fig. 2(b) shows agreement between the predicted conversion history of the particle and the experimental results. The conversion extent was defined as the mass ratio of the products to the virgin biomass. Since the predicted temperature

distributions in the particle are lower than the experiment during pyrolysis, the reaction rate is also lower, which leads to a lower conversion rate from 150 s to 300 s. After that, the value of the predicted conversion extent reaches the same level as in the experiment.

Table 1. Values of parameters used in the numerical simulation [14]

Property	Value
Thermal conductivity of wood, λ_w , $\text{W m}^{-1} \text{K}^{-1}$	0.1256
Thermal conductivity of charcoal, λ_c , $\text{W m}^{-1} \text{K}^{-1}$	0.0837
Density of wood bulk, ρ_w , kg m^{-3}	500
Density of charcoal bulk, ρ_c , kg m^{-3}	300
Specific heat capacity of wood, C_{pw} , $\text{J kg}^{-1} \text{K}^{-1}$	1670
Specific heat capacity of charcoal, C_{pc} , $\text{J kg}^{-1} \text{K}^{-1}$	1000
Activation energy, E , J mol^{-1}	67500
Pre-exponential factor, A , s^{-1}	2500

As shown in Fig. 3, the predicted conversion histories at different conditions agree with the experimental results. Note that “d0.6 T780” corresponds to the case of particle diameter 0.6 cm and reactor temperature 780 K. Overall, the present LBM model is capable of predicting the conversion history of biomass particles under various pyrolysis conditions.

3.2 Validation 2: Experiment by Park et al. [15]

Park et al. [15] studied the mass and heat transfer processes during wood pyrolysis. In their experiment, wood spheres with a 25.4 mm diameter were pyrolyzed at temperatures in the range from 638 K to 879 K. The temperature and mass losses of the biomass particle were measured in the experiment. The authors also proposed a reaction model for wood pyrolysis [15]. Since the development of chemical reaction mechanisms was not the focus of the present study, we utilized the same kinetic model and parameters as suggested by Park et al. [15]. The reaction pathways are shown in Fig. 4. In the simulation, boundary conditions with velocity inlet and

pressure outlet were used, and the wall temperature were kept constant. The initial porosity of biomass particle was 0.4.

Three competitive primary reactions describe the generation of tar, syngas, and intermediate solid from virgin biomass. Then, the intermediate solid is further converted to char, while the cracking of tar leads to the forming of the secondary syngas and char via two competitive reactions. The corresponding kinetic parameters are listed in Table 2. The reactor and gas temperatures used in the experiments are listed in Table 3, while the physical properties of biomass particles are shown in Table 4. By applying the conservation equations Eq. (5)–(7), the following can be obtained.

$$\text{Virgin wood:} \quad \frac{\partial \rho_b}{\partial t} = \omega_b = -(k_t + k_{syn} + k_{is})\rho_b \quad (21)$$

$$\text{Intermediate solid:} \quad \frac{\partial \rho_{is}}{\partial t} = \omega_{is} = k_{is}\rho_b - k_c\rho_{is} \quad (22)$$

$$\text{Char:} \quad \frac{\partial \rho_c}{\partial t} = \omega_c = k_c\rho_{is} + k_{c2}\rho_t \quad (23)$$

$$\text{Tar:} \quad \frac{\partial(\varepsilon\rho_t)}{\partial t} + \nabla \cdot (\rho_t \mathbf{u}_f) = \omega_t = k_t\rho_b - (k_{c2} + k_{syn2})\rho_t \quad (24)$$

$$\text{Syngas:} \quad \frac{\partial(\varepsilon\rho_{syn})}{\partial t} + \nabla \cdot (\rho_{syn} \mathbf{u}_f) = \omega_{syn} = k_{syn}\rho_b + k_{syn2}\rho_t \quad (25)$$

$$\begin{aligned} \text{Temperature:} \quad & (\rho_b C_{Pw} + \rho_{is} C_{Pw} + \rho_c C_{Pc} + \varepsilon\rho_t C_{Pt} + \varepsilon\rho_{syn} C_{Psyn}) \frac{\partial T_s}{\partial t} + (\rho_t C_{Pt} \\ & + \rho_{syn} C_{Psyn}) \nabla \cdot (T_s \mathbf{u}_f) = \lambda_s \nabla^2 T_s + Q \end{aligned} \quad (26)$$

$$Q = -(k_t \Delta h_t + k_{syn} \Delta h_{syn} + k_{is} \Delta h_{is})\rho_b - k_c \Delta h_c \rho_{is} - (k_{c2} \Delta h_{c2} + k_{syn2} \Delta h_{syn2})\rho_t \quad (27)$$

Table 2. Kinetic parameters used in the model [15]

Reaction	t	syn	is	c	c2	syn2
A_i (s^{-1})	1.08×10^{10}	4.38×10^9	3.75×10^6	1.38×10^{10}	1.0×10^5	4.28×10^6
E_i (J/mol)	148000	152700	111700	161000	108000	108000
Δh_i (kJ/kg)	80	80	80	-300	-42	-42

Table 3. Reactor and gas temperatures (K) [15]

T_{wall}	638	688	736	783	831	879
T_{in}	585	635	670	720	770	820

Table 4. Physical properties [12, 15]

Property	Value
Density of wood, ρ_w	630 (kg/m^3)
Specific heat capacity of wood, C_{Pw}	$1500 + 1.0T$ (J/kg K)
Specific heat capacity of char, C_{Pc}	$420 + 2.09T + 6.85 \times 10^{-4}T^2$ (J/kg K)
Specific heat capacity of tar, C_{Pt}	$-100 + 4.4T - 1.57 \times 10^{-3}T^2$ (J/kg K)
Specific heat capacity of syngas, C_{Psyn}	$770 + 0.629T - 1.91 \times 10^{-4}T^2$ (J/kg K)
Thermal conductivity of wood, λ_w	0.20487 (W/m K)
Thermal conductivity of char, λ_c	0.0937 (W/m K)
Thermal conductivity of gas, λ_g	0.0258 (W/m K)
Viscosity of gas, μ	3.0×10^{-5} (kg/m s)
Universal gas constant, R	8.314 (J/ mol K)
Stefan-Boltzmann constant σ	5.67×10^{-8} (W/m ² K ⁴)
Pore diameter d	$5 \times 10^{-5}(1-\eta) + 1 \times 10^{-4}\eta$ (m)

In the original study [15], the effective thermal conductivity was used to consider the compositional change and radiative heat transfer during the conversion of the biomass particle.

$$\lambda_{eff} = (1-\eta)\lambda_w + \eta\lambda_c + \varepsilon\lambda_g + 13.5\sigma T^3 d \quad (28)$$

σ and d are the Stefan-Boltzmann constant and pore diameter, respectively. The specific heat capacity was calculated as a function of temperature, as shown in Table 4. Constant thermal conductivity and specific heat were adopted in a number of studies to simplify the calculation [21-23]. As shown in the energy equation, Eq. (26), these thermophysical properties are important in the heat transfer process and they need to be evaluated carefully. In this work, several modeling approaches were tested to characterize the effect of thermal conductivity and specific heat on the prediction of biomass fast pyrolysis, as shown in Table 5.

Table 5. Test cases using different thermophysical properties

	Thermal conductivity	Specific heat capacity
Case 1	$\lambda = \lambda_w + \varepsilon\lambda_g$	Constant C_{Pave}
Case 2	$\lambda = (1-\eta)\lambda_w + \eta\lambda_c + \varepsilon\lambda_g$	Constant C_{Pave}
Case 3	$\lambda = (1-\eta)\lambda_w + \eta\lambda_c + \varepsilon\lambda_g + 13.5\sigma T^3 d$	Constant C_{Pave}
Case 4	$\lambda = (1-\eta)\lambda_w + \eta\lambda_c + \varepsilon\lambda_g$	Variable C_P as listed in Table 4
Case 5	$\lambda = (1-\eta)\lambda_w + \eta\lambda_c + \varepsilon\lambda_g + 13.5\sigma T^3 d$	Variable C_P as listed in Table 4

Here, the averaged value for specific heat capacity is defined as

$$C_{Pave} = \frac{\int_{T_s}^{T_{wall}} C_P dT}{T_{wall} - T_s} \quad (29)$$

where T_s is the initial temperature of the biomass particle and T_{wall} is the wall temperature. In

Case 1, the thermal conductivity consists of a constant and a variable, considering the change in

gas properties, while the specific heat is constant. In Case 2, the thermal conductivity accounts for the compositional change of the particle (from biomass to char) and the gas, while the specific heat is constant. In Case 3, the contribution of radiative heat transfer was considered. Case 4 and 5 are parallel to Case 2 and 3, respectively, while the specific heat is a function of temperature. The case of 783 K (T_{wall} 783K and T_{in} 720K) was chosen for the sensitivity study and the results are discussed here.

The predicted solid mass fractions are compared with the experimental data in Fig. 5. The solid mass fraction is defined as the mass ratio of the solid phase (sum of unreacted biomass, intermediate solid, and char) to the virgin biomass. It can be seen that, the predicted solid mass fraction decreased more slowly than the experimental data. This is the result of relatively slow heat transfer. As shown in Fig. 6, the predicted center temperature increased more slowly than the experimental data. When comparing the five cases, one can find that the center temperature increased the fastest in Case 1 and most slowly in Case 4. This is because in Case 1 the thermal conductivity was almost constant and the heat capacity was constant, while in Case 4 the thermal conductivity varied with composition and the heat capacity also varied with temperature. By comparing Case 1, 2 and 3, we can find that the thermal conductivity decreased during the conversion from biomass to char (Table 5), thus heat transfer was slower in Case 2 and 3. The inclusion of radiative heat transfer in the thermal conductivity has led to an increased center temperature. From the comparisons between Case 2 and 3, as well as Case 4 and 5, it is seen that the center temperature increased more slowly when the heat capacity was a function of temperature.

Significant discrepancies between experiments and predictions are seen in Fig. 5 and 6. One possible reason is that the present simulation used a 2D approach, in which the particle was

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4 treated more like an infinitely long cylinder rather than a sphere. Therefore, the present
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6 numerical approach was improved by considering the 3D effect in the 2D modeling, namely the
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8 heat transfer in the “third direction.”
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12 Details of the numerical method is described in the Supplemental Material. In the new
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14 approach, a modified cylindrical coordinate was used, resulting in an additional term in the
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16 energy equation [45]. This new term accounts for the heat transfer from the direction
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18 perpendicular to the 2D simulation domain. As a result, the new term considers the heating that
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20 is not present in the original 2D approach because of the assumption of an infinitely long
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22 cylinder. The numerical results using the new approach are shown in Fig. 7 and 8. The
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24 agreement between predictions and measurements is greatly improved.
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31 Results of Case 1, 3 and 5 are shown here for discussion. Case 5 has the most complex
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33 formulations for thermal conductivity and heat capacity, and it is of interest to explore the effects
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35 of these complex formations on simulation results.
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40 The predicted solid mass fractions are compared with the experimental data at different
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42 reactor temperature conditions in Fig. 9. Good levels of agreement can be seen, except for the
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44 case of 638 K. As shown in Fig. 10, the predicted history of the center temperature also agrees
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46 with the experimental data. In Case 5, the center temperature increased more slowly and the peak
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48 temperature occurred later than the measured data. Overall, the results indicate that the use of
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50 constant heat capacity is acceptable and the use of constant thermal conductivity can also be
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52 acceptable at low reactor temperature conditions. In the following, only the results of Case 5 will
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54 be shown, while the results of Case 3 and 1 can be found in the Supplemental Materials.
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The temporal evolutions of the pyrolysis product yields at different reactor temperatures are shown in Fig. 11. The final product yields predicted by the present model were found to be very close to the numerical results of Park et al. [15]. As the temperature increased in the particle, the reaction rates were accelerated and the product yields increased rapidly at the beginning. It can be seen that, for the case of 783K, the tar and syngas yields reached the maximum at 380 s, while the char yield continued to increase until around 400 s. After that, the product yields did not change as the pyrolysis process was completed and only char remained in the solid phase. A similar process can be observed in the case of 688 K.

In general, the temperature in a fast pyrolysis reactor was around 500 °C, thus the results of the 783 K case were analyzed in detail. Fig. 12 shows the distributions of gas velocity and temperature in the reactor at 100 s. The black circle represents the surface of the particle. Fig. 12(a) shows that the maximum velocity occurred in the area between the particle and the wall, due to the conservation of mass. Since biomass is porous, the gas flows through the particle, as seen from the streamlines in Fig. 12(a). The gas velocity near the particle affected the convective heat transfer between gas and particle, and consequently affected the temperature distribution in the particle, as shown in Fig. 12(b). In the experiment, the temperature inside the particle was measured in the radial direction from the center to the surface point 1, which was perpendicular to the inlet gas velocity, as shown in Fig. 12(b). Since the gas came from the left, it is also interesting to reveal the temperature distributions in the radial direction, from the center to the front (surface point 2), and from the center to the back (surface point 3), which were not mentioned in the original paper [15].

The temperature distributions along these three radial directions are shown in Fig. 13(a). It can be seen that, at radial positions smaller than 0.7, the temperatures at radial direction 2 and

radial direction 1 were slightly higher than radial direction 3, while at radial locations larger than 0.7, temperatures at radial direction 1 increased faster than radial direction 2. Radial direction 3 had the lowest temperature. The distribution of temperature can be explained by the effect of gas flow around the particle. Since the inlet gas temperature was lower than the reactor wall temperature, as shown in Table 3, the gas was heated when flowing through the reactor. From Fig. 12(b), it is found that the gas temperatures near particle surface point 1 was higher than those near point 2 and 3, resulting in higher temperature at radial direction 1 at radial position larger than 0.7.

Different temperatures produced different conversion rates. High temperature is favorable for high reaction rate, causing more biomass to be converted. The fractions of solid mass (including raw biomass, intermediate solid, and char) at different radial directions at 100 s are presented in Fig. 13(b). At the radial position larger than 0.7, the solid mass fraction decreased as temperature increased because more biomass was decomposed. Radial direction 1 had the minimum solid mass fraction while radial direction 3 had the maximum solid mass fraction. By comparing Fig. 13(a) and (b), it is seen that, at 100 s, the solid mass fractions at radial positions smaller than 0.7 were nearly unchanged, indicating biomass will not be converted when temperature is below 550 K.

The evolution of the surface temperatures at different radial directions are presented in Fig. 14. It can be seen that the surface temperatures increased rapidly and finally reached a steady state, which was between the gas temperature and the wall temperature. The surface temperature at point 2 increased faster than point 3 before 100 s, but then increased more slowly than point 1 and 3. This is due to the heat transfer condition at the front of the particle (point 2) where the gas temperature was lower, while the gas temperature was higher near point 1 and 3, due to heating

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4 by the reactor wall. At the beginning, heat transfer at the front of the particle was more effective;
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6 after 100 s, the convective heat transfer became more dominant. It can be found that the
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8 temperature at the front of the particle is strongly affected by the inlet gas temperature, as the
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10 final surface temperature at point 2 was close to the inlet gas temperature. On the other hand, the
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12 final surface temperatures at points 1 and 3 were close to the wall temperature. These results
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14 indicate that the temperature within the particle is affected by both the convective heat transfer
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16 from the gas and the heat conduction from the reactor wall. Therefore, using a constant
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18 temperature or heat flux at the particle surface by conventional 1D models will introduce
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20 inaccuracy. The intra-particle transport phenomena are highly sensitive to the surrounding flow
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22 conditions, which change rapidly in the reactor during fast pyrolysis. In order to simulate
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24 biomass fast pyrolysis at the particle scale more accurately, the intra-particle transport needs to
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26 be solved by coupling the particle and the surrounding gas flow, as was done in this study.
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34 The in-particle temperature distribution has a pronounced effect on the conversion of the
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36 biomass particle since chemical kinetics is driven by temperature. The gas flow also affects the
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38 transport of the resulting gaseous products. Chemical reactions start from the outer part of the
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40 particle, because of the higher temperature, and propagate inward. Fig. 15 shows the density
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42 distributions of raw biomass, tar, syngas, and char at 300 s, when the conversion process is
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44 mostly completed (Fig. 11(b)).
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48 As shown, chemical reactions have taken place in the entire particle. At 300 s, only a small
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50 portion of raw biomass remains near the center of the particle, where the temperature is the
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52 lowest. Much of tar and syngas have been generated, and these gaseous products are transported
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54 downstream. High concentrations of tar and syngas exist at the inner part of the particle at this
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56 time. Char is formed at the outer part of the particle where the conversion process first starts. As
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the conversion process progresses, tar and intermediate solid biomass will be converted to char. Eventually char will be the only solid in the particle, while tar and syngas will be transported downstream.

3.3 Sensitivity analysis

In this study, the physical and chemical parameters used in the model were adopted from the original experimental paper [15]. A sensitivity study was conducted here to characterize the effects of several important parameters on simulation results. These parameters include the physical property (density and porosity) and heat of reactions, as shown in Table 6. The conversion time was defined as the time when pyrolysis reached 95% conversion extent. The 783 K condition of Case 5 (Table 5) was discussed and shown here. Results of Case 3 and 1 are documented in the Supplemental Material.

In conducting the sensitivity study, one parameter was varied by $\pm 50\%$ from its original value. The sensitivity, S , was defined in the following [44].

$$S = \frac{\% \text{ change in conversion time (product yield)}}{\% \text{ change in the specified variable}} \quad (30)$$

The relative conversion time (compared with the baseline result) with respect to the change of each parameter is shown in Fig. 16. The conversion time increased with the increase in particle density. This is because increase in density will slow down heat transfer, as shown in the energy equation, Eq. (26). More energy is needed for the pyrolysis of high-density particles. Increasing porosity slightly decreased the conversion time, which is equivalent to increasing the thermal conductivity, according to Eq. (28).

Regarding the heat of reaction, the three primary reactions (k_t , k_{syn} , k_{is}) are endothermic, while the conversion from intermediate solid to char (k_c) is exothermic. Therefore, increasing the Δh_c will decrease the conversion time, since more energy will be released by the reaction k_c . On the contrary, the conversion time will increase with the increase in Δh_t , Δh_{syn} and Δh_{is} , because more energy is needed for these reactions. The effect of Δh_c is more significant compared to Δh_t , Δh_{syn} and Δh_{is} . Among the three endothermic reactions (k_t , k_{syn} , k_{is}), the change in conversion time for Δh_t is larger than Δh_{syn} and Δh_{is} . Although the value of Δh_t , Δh_{syn} and Δh_{is} are the same, the reaction rate of k_t is higher than k_{syn} or k_{is} , as seen in Table 2.

Results of the sensitivity, S , calculated by Eq. (30) are listed in Table 6. It can be seen that density is the most significant variable, while porosity is the least. The sensitivity of the product yield is smaller than the conversion time. The change of the char yield is similar to the conversion time, while the change of tar and syngas yield is opposite. This is because tar and syngas yields are favored at high temperature (lower conversion time), while more char will be generated at low temperature (higher conversion time). The only exception is the porosity. Increasing porosity will increase the thermal conductivity, causing the conversion time to decrease, as discussed earlier. However, the product yield of tar and syngas will also decrease as porosity increases. This can be explained by the mass equation, Eq. (24) and (25).

Table 6. Results of sensitivity study

Parameter varied	Conversion time	Tar yield	Syngas yield	Char yield
density	0.82	-0.04	-0.06	0.15
porosity	-0.01	-0.0002	-0.0005	0.001

Δh_c	-0.27	0.01	0.03	-0.06
Δh_{is}	0.05	-0.003	-0.005	0.01
Δh_{syn}	0.03	-0.002	-0.003	0.007
Δh_t	0.16	-0.01	-0.02	0.04

3.4 Effect of particle shape

As Ciesielski et al. [33] discussed in their paper, the results of using aspherical particle models were different from those of using spherical particle models. It should be noted that, in their paper, only transport of heat and mass were simulated without chemical reactions. To investigate the effect of particle shape on biomass fast pyrolysis, the results using an elliptic particle, with three different orientations to the inlet gas velocity, were compared with those of the circular particle and spherical particle. The aspect ratio of the elliptical particle was 4:1, which was close to the elliptical particles used in the experiment [33]. This aspect ratio was measured for the biomass particle after milling. Note that the major radius and minor radius were adjusted such that the cross-section area of the elliptic particle was the same as that of the circular particle. In addition, the flow area between the particle and the reactor wall was also kept the same. The distributions of gas velocity and temperature at 100 s, for different configurations, are presented in Fig. 17. The parameters are based on those of Case 5 in Table 2. Results based on Case 3 and 1 are shown in the Supplemental Material.

As shown in Fig. 17(a), for the elliptic particle with the major axis parallel to the wall, the predicted center temperature is higher than that of the circular particle shown in Fig. 12(b). The results using the elliptic particle with the major axis perpendicular to the wall are shown in Fig. 17(b). It can be seen that the maximum gas velocity is higher than that using the circular particle because of the increased obstruction of the flow area by the particle. The center temperature is

also higher than that using the circular particle. Fig. 17(c) illustrates the results of an elliptic particle with the major axis slanted at 45 degrees. The maximum velocity is also higher than that using the circular particle, while the center temperature is still higher than that of circular particle. The higher center temperature of elliptic particles can be explained as follows: Compared to the circular shape, an elliptic particle is flatter and thermally thinner, and therefore, heat can penetrate into the particle more easily; and the perimeter of an ellipse is larger than that of a circle under the same area, which is beneficial for the convective heat transfer from the surrounding gas.

Fig. 18 shows the evolution of the center temperatures of the particles with different shapes. It can be seen that the center temperature of an elliptic particle, regardless of its orientation, increases faster than that of a circular particle. More specifically, the center temperature of an elliptic particle placed horizontally increases the fastest, while the center temperature of an elliptic particle placed vertically increases the slowest. These results can be explained by the combination of heat transfer from the gas and the reactor wall. For a horizontal elliptic particle, the gas temperature near the top and the bottom of the particle is higher than the gas temperature near the front, due to the heating by the reactor wall. Considering the surface area at the top and at the bottom, the convective heat transfer from the gas in these areas is stronger than the heat transfer condition at the front. As opposed to the horizontal elliptic particle, the effect of heat transfer at the front of a vertical elliptic particle is important to particle heating since the surface area is larger at the front. Since the gas temperature at the front of the particle is lower than the gas temperature near the top and the bottom, the center temperature of a vertical elliptic particle increases more slowly than that of a horizontal elliptic particle, as shown in Fig. 18. The center temperature of an elliptic particle slanted at 45 degrees lies between these two.

When compared to the results of the spherical particle (using the new model), the center temperature of the elliptic particle increased faster at the beginning (before 150 s) and only the horizontal elliptic particle had a higher center temperature than the spherical particle after 350 s.

It should be noted that, the final center temperature is related to the surface temperature, which is also affected by the particle shape, as shown in Fig. 18. Since the final surface temperature is between the gas temperature and the wall temperature, the final center temperature is also found to be within this range. However, the final center temperature may be different for different particles, sometimes being closer to the gas temperature and sometimes being closer to the wall temperature. Fig. 18 illustrates that the final center temperature of a horizontal elliptic particle is very close to the wall temperature, while the final center temperature of a vertical elliptic particle is closer to the gas temperature, which is determined by the surface area at the front to the surface areas at the top and the bottom. If the surface area is larger at the front, the effect of heat transfer with lower gas temperature will be more significant when compared to the convective heat transfer with higher gas temperature at the top and the bottom of the particle, which results in lower final center temperature. By contrast, if the surface areas at the top and the bottom are larger, the convective heat transfer with higher gas temperature will be stronger than the heat transfer at the front of the particle, leading to a higher final center temperature.

Table 7. Effect of particle shape on final product yields and conversion time

	Spherical particle	Circular particle	Elliptic particle slanted at 45 degrees	Horizontal elliptic particle	Vertical elliptic particle
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Tar yield	0.676	0.662	0.671	0.680	0.667
Syngas yield	0.109	0.105	0.108	0.110	0.106
Char yield	0.216	0.233	0.221	0.211	0.227
Conversion time (s)	369	559	407	323	458

The effect of the particle shape on the final product yields and conversion time are shown in Table 7. It can be seen that the horizontal elliptic particle requires the minimum conversion time, followed by the spherical particle, while the circular particle requires the maximum conversion time. Since the temperature in the elliptic particle increases faster than circular particle, more tar and syngas are produced, while less char is generated, as illustrated in Table 7. This can be explained by the fact that the char yield is favored at low pyrolysis temperature, while tar and syngas yields increase at high temperature.

3.5 Perspectives

This work demonstrates that the present LBM method is able to provide detailed information on the physicochemical evolution of biomass particles during fast pyrolysis. Future studies can include the simulation of a particle stream under fluidization conditions. The numerical results can be further analyzed to derive engineering models (e.g. gas-solid drag force and heat transfer models) for describing the overall heat and mass transfer of biomass particles under relevant operating conditions.

4 Conclusions

A numerical method based on the lattice Boltzmann method was used for simulating biomass fast pyrolysis at the particle scale. Both the intra-particle transport phenomena and the surrounding gas flow were considered. This work demonstrates that the current model is capable of accurately predicting the temperature and conversion of biomass particles under various

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4 pyrolysis conditions. The distributions of temperature and density are found to be non-uniform
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6 within the particle when undergoing fast pyrolysis. The non-uniform distributions of surface
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8 temperature suggest that constant temperature or heat flux assumptions, as often adopted by most
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10 numerical studies, will require modifications for more accurate results.
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15 Different formulations of thermophysical properties were tested. The results indicate that
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17 use of constant thermal conductivity and heat capacity is appropriate at low reactor temperature
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19 conditions. Sensitivity analysis shows that density is the most influential parameter while
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21 porosity is the least. Among the reactions studied, the heat of reaction, converting intermediate
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23 solid to char, is more dominate than the three primary reactions.
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28 To further investigate the effect of particle shape on biomass fast pyrolysis, a sensitivity
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30 study was conducted to examine the conversion time and final product yields. The particle with
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32 elliptic shape has higher center temperature and shorter conversion time than the circular particle.
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34 This is due to the larger surface area and the shorter distance from the center to the surface,
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36 compared to a circular particle. The results show that the center temperature of a horizontal
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38 elliptic particle increases the fastest, while the center temperature of a vertical elliptic particle
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40 increases the slowest. The effect of particle shape on the final center temperature can be
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42 explained by comparing the heat transfer condition at the front of the particle and the convective
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44 heat transfer at the top and the bottom of the particle. For an elliptic particle, more tar and syngas
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46 are produced while less char is generated, compared to a circular particle, because the
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48 temperature in the elliptic particle increases faster. These results indicate that the shape of the
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50 biomass particle should be considered when conducting biomass fast pyrolysis analysis.
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Acknowledgements

This work was supported by the National Science Foundation under the Grant Number EPS-1101284.

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Nomenclature

A	pre-exponential factor (s^{-1})	ρ	density (kg m^{-3})
C_P	specific heat capacity ($\text{J kg}^{-1} \text{K}^{-1}$)	τ	relaxation time
D	mass diffusion coefficient ($\text{m}^2 \text{s}^{-1}$)	ω	rate of production ($\text{kg m}^{-3} \text{s}^{-1}$)
E	activation energy (J mol^{-1})		
k	reaction rate (s^{-1})	Subscripts	
P	pressure (Pa)	ave	average value
Q	heat generation (W m^{-3})	c	char, primary char generation reaction
R	universal gas constant ($\text{J mol}^{-1} \text{K}^{-1}$)	c2	secondary char generation reaction
T	temperature (K)	f	fluid phase (tar, syngas)
t	time (s)	g	gas
U	gas velocity (m s^{-1})	i	specie i, lattice direction i
		is	intermediate solid
Greek letters		j	reaction j
α	thermal diffusivity ($\text{m}^2 \text{s}^{-1}$)	s	solid phase
ε	porosity	syn	syngas, primary syngas generation reaction
η	conversion extent of pyrolysis	syn2	secondary syngas generation reaction
Δh	heat of reaction (J kg^{-1})	t	tar, primary tar generation reaction
λ	thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)	T	temperature
μ	viscosity ($\text{kg m}^{-1} \text{s}^{-1}$)	w	wood
ν	kinematic viscosity ($\text{m}^2 \text{s}^{-1}$)	in	inlet condition

Figure Captions

Fig. 1. (a) Schematic of domain discretization by LBM (b) description of particle in lattice.

Fig. 2. Comparisons between experimental and numerical results ($d=2.2$ cm, $T_{\text{wall}}=753\text{K}$):
(a) radial temperature distribution at different times; (b) conversion history of biomass particle.

Fig. 3. Comparisons of conversion history between experimental and numerical results at
different conditions

Fig. 4. Reaction model used by Park et al. [15]

Fig. 5. Evolutions of measured and predicted solid mass fractions

Fig. 6. Evolutions of measured and predicted center temperatures of biomass particle

Fig. 7. Evolutions of measured and predicted solid mass fractions

Fig. 8. Evolutions of measured and predicted center temperatures of biomass particle

Fig. 9. Evolutions of measured and predicted solid mass fractions: (a) Case 5 (b) Case 3 (c)
Case 1

Fig. 10. Evolutions of measured and predicted center temperatures of biomass particle: (a) Case
5 (b) Case 3 (c) Case 1

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7 Fig. 11. Predicted temporal evolutions of pyrolysis product mass fractions, compared with the
8 literature data [15] at different reactor temperature: (a) 688K (b) 783K
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13 Fig. 12. Spatial distributions of (a) velocity and (b) temperature at 100 s
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20 directions at 100 s
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Figure-1-a

Figure-1-b

Figure-2-a

Figure-2-b

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Figure-8

Solid mass fraction

Figure-9-b

Figure-9-c

Figure-10-a

Figure-10-b

Figure-10-c

Figure-11-a

Figure-11-b

Figure-12-a

Figure-12-b

Figure-13-a

Figure-13-b

Figure-14

Figure-15-a

Figure-15-b

Figure-15-c

Figure-15-d

Figure-16

Figure-17-a-1

Figure-17-a-2

Figure-17-b-1

Figure-17-b-2

Figure-17-c-1

Figure-17-c-2

Figure-18