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Heat transfer corrected isothermal model for devolatilization of thermally-thick biomass particles

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Abstract: Isothermal model used in current computational fluid dynamic (CFD) model neglect the internal heat transfer during biomass devolatilization. This assumption is not reasonable for thermally-thick particles. To solve this issue, a heat transfer corrected isothermal model is introduced. In this model, two heat transfer corrected coefficients: H_T -correction of heat transfer and H_R -correction of reaction, are defined to cover the effects of internal heat transfer. A series of single biomass devolatilization case have been modeled to validate this model, the results show that devolatilization behaviors of both thermally-thick and thermally-thin particles are predicted reasonable by using heat transfer corrected model, while, isothermal model overestimate devolatilization rate and heating rate for thermally-thick particle. This model probably has better performance than isothermal model when it is coupled with CFD to model devolatilization of thermally-thick biomass particles.

1. Introduction

Eulerian-Eulerian methods and Eulerian-Lagrangian methods have been widely used in CFD simulations of biomass combustion and gasification in fluidized bed [1–4]. In these two approaches, the biomass particles are usually described by isothermal model which neglects the internal heat transfer in biomass particle. This model is simple and easy to couple with CFD software. However, it is only reasonable for the case with relatively small Biot Number (Bi) (e.g. $Bi < 1$) [5]. With the increase of Bi, the internal temperature gradient becomes large, resulting different reaction rate of radials position. Thus, the thermally-thick biomass particle devolatilization behaviour has significant difference as compared with isothermal described.

Pyle and Zaror [5] investigated wood pyrolysis under different Bi (0.25 ~66), showing that the isothermal assumption is reasonable for the case with $Bi < 1$. For particle with $Bi > 1$, isothermal model predicted shorter devolatilization time than the experimental data, due to the internal heat transfer gradually taking an dominated role when $Bi > 1$ [5]. Similar results also have been reported by Lu et al. [6] and Bharadwaj et al. [7]. Johansen et al. [8] also found that an isothermal model predicts shorter devolatilization time as compared with non-isothermal model. Since the internal heat transfer has significant effects on biomass devolatilization behaviours, non-isothermal model is favourably applied in this case [6,8–12].

Among the existing non-isothermal models, only a few of them have been implemented in CFD simulations. A detailed non-isothermal model, heat diffusion equation for an isotropic particle (a partial differential equation (PDE) at particle scale) has been implemented into Fluent® by using User Defined Function (UDF), as reported by Papadikis et al. [13–15]. The model can capture more detailed information of biomass particle (e.g, internal temperature gradient), as compared with traditional Eulerian-Eulerian methods. However, the simulation slows down significantly, due to the extremely high amount of memory allocation when coupled with detailed non-isothermal model [14]. In other way, Thunman et al. [16] proposed a simplified non-isothermal model. In their model, wood particle was divided into for different layers: moist

wood, dry wood, char, and core layer, and only ordinary differential equation (ODE) are required to solve. This model is investigated a lot in fixed bed [11,17,18], but it is rarely reported in fluidized bed.

As compared with non-isothermal model, isothermal model is still the most applicable method to simulate fluidized bed [19–21]. However, it is not reasonable to use to simulate the devolatilization of thermally-thick particle (e.g. $Bi > 1.0$). To modify the isothermal model, Johansen et al. [3] modified the kinetic parameters by fitting CFD modelling non-reacting biomass particle temperature history versus experimental conversion, instead of using kinetic parameters from single particle pyrolysis experiment. In this way, CFD modelling performance is improved. Simone et al. [22] also adopted similar method to simulate biomass devolatilization in a drop tube reactor. However, several CFD simulations are required to obtain appropriate kinetic parameters, it is time-consuming, and the derived kinetic parameters should be updated with operating conditions (e.g. Experimental setup, gas velocity).

In this paper, a heat transfer corrected model is introduced to model thermally-thick biomass devolatilization in fluidized beds. The isothermal model used in CFD simulations is modified both by a heat-transfer correction coefficient for heat flux, H_T , and a correction for reaction rate, H_R . Those parameters were determined from non-isothermal model under different heat convection coefficients, thus, it can be used in arbitrary operating conditions with constant particle size and gas/wall temperature as well as arbitrary experimental setup. A series of cases studies have been carried in this study to verify the developed model.

2. Model derivation

According to uniform temperature assumptions of isothermal model, the heat transfer between particle and fluid is described as below:

$$q_{iso} = h_c A_p (T_g - T_p) + \xi \sigma A_p (T_w^4 - T_p^4) \quad (1)$$

However, particle surface temperature (T_{surf}) is larger than particle temperature (T_p), during heating thermally-thick particle. The heat transfer between particle and fluid should be described as in non-isothermal model:

$$q_{non-iso} = h_c A_p (T_g - T_{surf}) + \xi \sigma A_p (T_w^4 - T_{surf}^4) \quad (2)$$

Since T_{surf} is larger than T_p , isothermal model overestimates the heat transfer rate between gas and solid. Moreover, radial profiles of volatiles or water mass fraction is also not uniform during biomass devolatilization, resulting significant different behaviours of what isothermal model predicted. Therefore, hereby two heat transfer coefficients are defined, H_T , accounting for the heating flux difference between isothermal model and non-isothermal model, and H_R , accounting for the reaction rate constant difference between isothermal model and non-isothermal model. Those two factors are determined as follows:

$$H_T = \frac{q_{non-iso}}{q_{iso}} = \frac{h_c (T_g - T_p) + \xi \sigma (T_w^4 - T_p^4)}{h_c (T_g - T_{surf}) + \xi \sigma (T_w^4 - T_{surf}^4)} \quad (3)$$

$$H_R = \frac{R_{non-iso}}{R_{iso}} = \frac{\int_0^R 4\pi r^2 f \rho_B dr}{f \rho_B V_p} \quad (4)$$

Where f is arbitrary reaction model (Drying model and devolatilization model) to describe biomass devolatilization.

According to Eq.3, H_T is a function of (T_p, h_c, T_{surf}) . Since T_p and T_{surf} is constrained by Fourier's law [8], H_T and H_R are simplified as a binary function of (T_p, h_c) . H_T and H_R are possible used in different operating conditions (e.g. gas velocity and pressure) under same gas/wall temperature, because such changes are represents by h_c .

For the ease of using in CFD software, a dimensionless temperature (θ) are defined as below.

$$\theta = \frac{T_p - T_{p0}}{\max(T_w, T_g) - T_{p0}} \quad (5)$$

After arrangement, H_T and H_R are expressed as binary function of (θ, h_c) . The heat transfer corrected isothermal model are described as follows:

Heat balance:

$$C_p m_p \frac{dT_p}{dt} = H_T (h_c A_p (T_g - T_p) + \xi \sigma A_p (T_w^4 - T_p^4)) - R_p Q_R \quad (6)$$

Mass balance:

$$R_p = H_R R_{iso} \quad (7)$$

H_T and H_R are determined by using following solving scheme:

- (1) With given input parameters: gas/wall temperature (T_g, T_w) and particle diameter (d_p), traverse over all possible values of h_c from $h_{c,min}$ to $h_{c,max}$. $h_{c,min}$ and $h_{c,max}$ covers all possible values during devolatilization.
- (2) Solve the non-isothermal model to obtain particle average temperature (T_p), average biomass conversion (X_p), heating transfer rate ($q_{non-iso}$), and reaction rate ($R_{non-iso}$). The inputments of this step are drying model, devolatilization kinetic model, algebraic equations to determine particle properties, and heat of water evaporation biomass devolatilization;
- (3) Calculate dimensionless temperature θ as Eq. 5;
- (4) Calculate H_T and H_R based on Eq. 3 and Eq. 4;
- (5) Save and output H_T, H_R, θ and h_c , those data either used by fitting H_T and H_R as binary function of (θ, h_c) or linear interpolation.

Fig. 1 shows the heat transfer corrected coefficients, $H_T(a)$ and H_R , of a thermally-thick biomass particle ($d_p = 10$ mm). For H_T , it decreases with the increase of dimensionless temperature (θ), due to $T_{surf} < T_p$. At the two ends, $\theta=0$ and $\theta=1$, H_T trends to 1. H_T declines with the increase of heat convection coefficients (h_c), because large h_c leads to large internal temperature gradient.

For H_R , H_R is increased firstly with the increase of θ , and then decreased to lower than 1, and finally increase back to 1. At the two ends, H_R also corresponds to 1. H_R is in a range of $10^{-4} \sim 10^{10}$, it indicates that non-isothermal model predicts significant difference biomass devolatilization behaviors, as compared with isothermal model. H_T and H_R determined from samll particle ($d_p=0.1$, $Bi=0.019\sim 0.274$). Both H_T and H_R is close to 1 with a range of h_c from 1 to $500 \text{ W/m}^2/\text{K}$, it means that biomass devolatilization behaviors are kinetic controlled, instead of heat transfer.

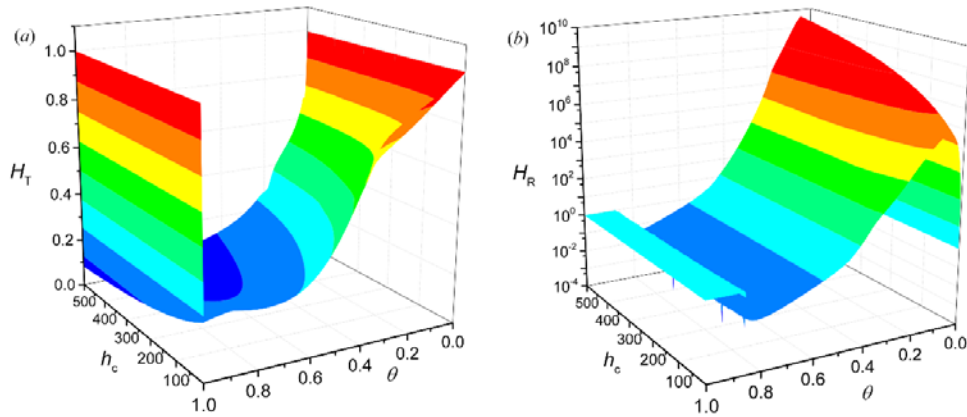


Figure 1 The heat transfer corrected coefficients (H_T and H_R) for isothermal model (Gas: N_2 , cylinder particle, $\rho_B=615 \text{ kg/m}^3$, 78% volatiles, 0% moisture, $T_g=T_w=1059 \text{ K}$, $d_p=10 \text{ mm}$, $h_c=1\sim 500 \text{ W/m}^2/\text{K}$, $Bi=1.9\sim 27.4$, physical properties are described in section 3)

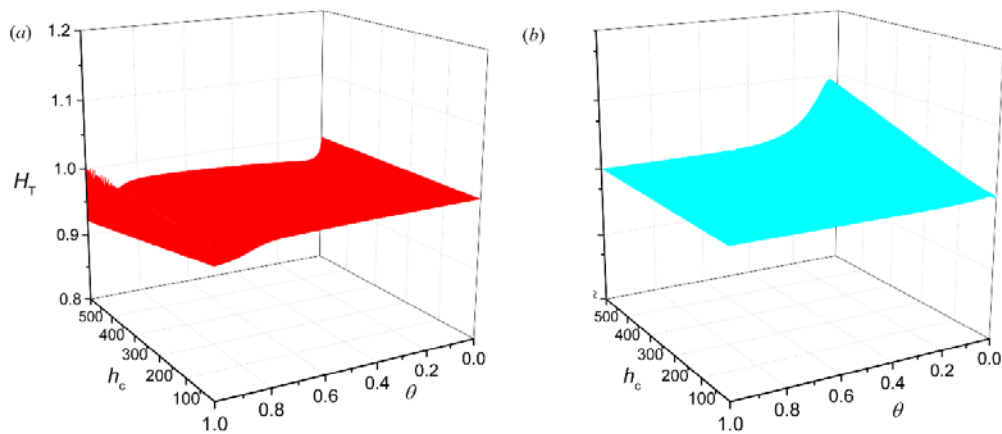


Figure 2. The heat transfer corrected coefficients (H_T and H_R) for isothermal model (Gas: N_2 , cylinder particle, $\rho_B=615 \text{ kg/m}^3$, 78% volatiles, 0% moisture, $T_g=T_w=1059 \text{ K}$, $d_p=0.1 \text{ mm}$, $h_c=1\sim 500$, $Bi=0.019\sim 0.274$, physical properties are described in section 3).

3. Model validation

3.1. Experimental setups and operating conditions

Table 1 shows the particle size, operating conditions and its references. For the case of small particle, the reactor description and experimental procedure can be found in reference [3]. The

bulk gas flow is laminar flow with slip velocity ~ 5 m/s, and operating temperature is 1405-1667 K. For the case of middle size particle, the experiments were performed in a single particle combustor (SPC) as described in [23]. The devolatilization time is estimated from the images of volatiles combustion flames. For the case of large particle size, two different experimental data (Lu et al. [6] and Alves and Figueiredo [10]) are used to evaluate different models. The details of proximate analysis of each case are shown in Table 2.

Table 1. Overviews of the experimental setup and operating conditions used for model validation

Cases	Set up	Particle size (mm)	Density (kg/m ³) (dry based)	Gas/wall Temperature (K)	Ref.
Small	Entrained flow reactor	0.063~0.09	1180	1405~1667	[8]
Middle	Single particle combustor	1.31-3.0	544	1473~1873	[23]
Large	Single particle combustor	9.5	580	1050~1276	[6]
	Vertical steel reactor	~ 18.5	615	573~1059	[10]

Table 2. Overviews of the experimental setup and operating conditions used for model validation

Parameter	Small [8]	Middle [23]	Large	
			Single particle combustor [6]	Vertical steel reactor [10]
Moisture (dwt%)	0.0	10.3	6.38	0.0
Volatiles (dwt%)	91.9	85.3	90	78
Char (dwt%)	7.90	14.3	9.5	22
Ash (dwt%)	0.2	0.4	0.5	0.0

3.2. Thermo-physical properties model

The effective heat conductivity (λ_{eff}) of is express as the sum of the conduction in the solid phase (λ_p), gas phase ($\varepsilon_g \lambda_g$), liquid moisture ($\varepsilon_w \lambda_w$), and radiative (λ_r) [24]. The λ_p is correlated as a function of volatiles conversion. λ_r is correlated from previous references [8,24]. particle pore diameter is assumed to vary linearly with the conversion [8]. The heat capacity of wood and char are found from references [6,8,25]. The heat required for water vaporization is the sum of heat of vaporization and desorption [8,26]. The total particle shrinking is assumed to 0.2 during drying and pyrolysis and it assumed vary linearly with total conversion. Both particle porosity and density are correlated as function of shrinkage coefficients (φ) and species contents. Emissivity is 0.85. More details of above thermo-physical properties can be found in reference [8]. The heat convection coefficient of spherical particle is determined by well-known Ranz and Marshall equation [27], while the correlation of Churchill and Bernstein is adopted for cylinder particle [28].

3.3. Drying and pyrolysis model

The drying of the particle is described by a first order kinetic model:

$$\frac{d\rho_w}{dt} = k_w (Y_{w0} - Y_w) \rho_B \quad (8)$$

The rate constant k_w is expressed as Arrhenius expression:

$$k_w = k_{w0} \exp\left(\frac{-Ea_w}{RT}\right) \quad (9)$$

The kinetic data of Bryden et al.[29], where $k_{w0}=5.13 \times 10^{10}$ and $Ea_w=88$ KJ/mol is used in this study. This kinetic data were also used by other references[6,30].

For wood devolatilization kinetic, a two-step semi-global reaction mechanism is adopted in this study. The kinetic data were derived by Wagenaar et al.[31] and has been successfully used to simulate both small and large particle [6,8,19]. Following by Johansen et al.[3], The devolatilization rate is expressed as below:

$$\frac{dY_v}{dt} = (k_v + \min(k_t, k_{rv}))(Y_{v0} - Y_v) \quad (10)$$

The rate constants are expressed as Arrhenius expression, the kinetic parameters are listed in Table 3. The heat of wood devolatilization is in range of -400 to 500 kJ/kg [11,32]. In this study, it is assumed to be 418 kJ/kg for most the cases, this value has been validated by Lu et al.[6]. For the cases of Alves and Figueiredo [10], the heat of devolatilization are calculated based on their kinetic data, it is 95.76 kJ/kg.

Table 3. Kinetic parameters of two-step semi-global reaction mechanism

Kinetic parameters	Unit	Constant
k_{v0}	s^{-1}	1.11×10^{11}
Ea_v	KJ/mol	177
k_{t0}	s^{-1}	9.29×10^9
Ea_t	KJ/mol	149
k_{rv0}	s^{-1}	4.28×10^6
Ea_{rv}	KJ/mol	107.5

3.4. Results and dicussions

Fig. 3 shows a comparison between measurements and the devitalization profiles of pinewood predicted by different models at 1405 and 1667 K of small particle ($d_p=78.9 \mu m$, $Bi \sim 1$). Non-isothermal and this work predict similar results, and isothermal model predicts slightly faster conversion rate than the other two models. All models show a good agreement with the experimental model. Those results indicate that both isothermal and non-isothermal model is

reasonably used to predict the devitalization behavior of small particles. Similar results were also reported by other researches [5,7].

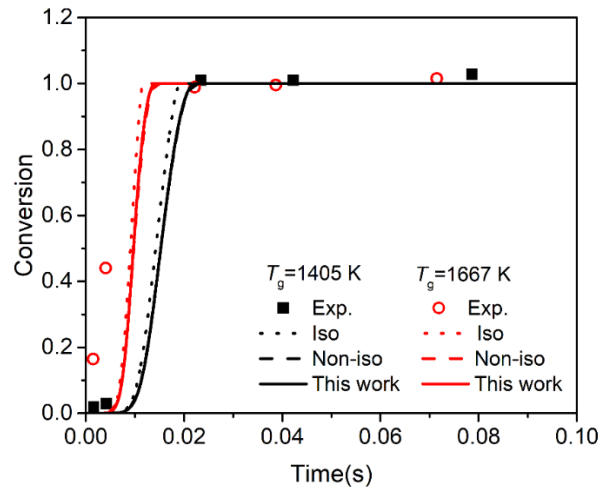


Figure 3. Comparison of the conversion history of experimental data and the results predicted by different models

Fig. 4 shows the devitalization time (99% conversion) predicted by different models and its comparison with the experimental data from Momeni et al.[23]. Fig. 4 (a) show the effects of particle size (the case of $d_p=3$ mm is spherical particle, the others are cylinder particles, more details can be found in Momeni et al. [23]). The particle is devitalized at $T_g=T_w=1473$ K with 20% oxygen. Both non-isothermal model and this work predict similar results, and the results predicted by those two models are in good agreement with experimental data, while isothermal model is underestimate the devolatilization time for all cases. The deviation between different isothermal model and other two models are decreased with the decrease of particle size (Bi is decreased from a range of 2.7~9.5 to 1~4). Fig. 4 (b) presents the devitalization time under different bulk gas temperatures ($d_p=1.65$ mm). Similar to Fig. 4 (a), isothermal model predict shortest devitalization time, while non-isothermal model and this work predicts similar results.

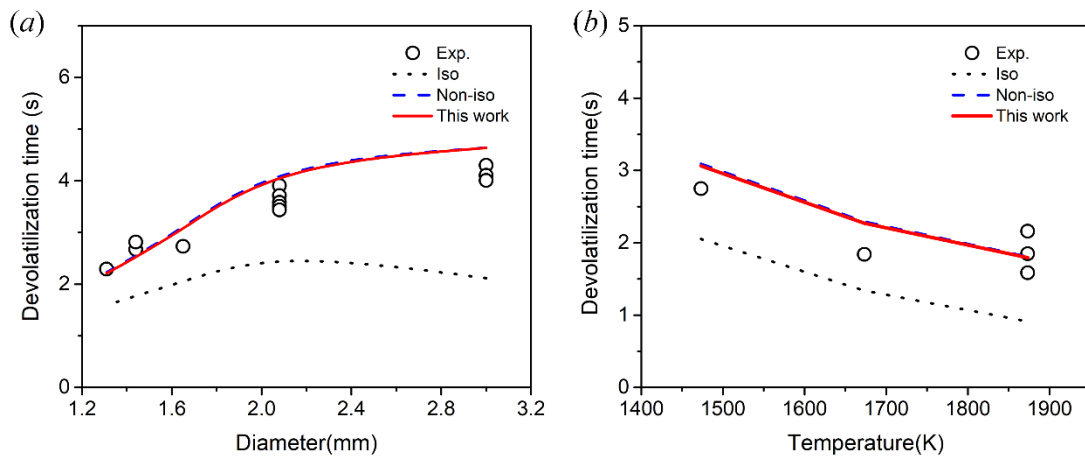


Figure 4. The devitalization time of wood particle predicted by different models and its comparison with experimental data: (a) the effects of particle size, (b) the effects of bulk gas temperature.

Fig. 5 (a) show the conversion predicted by different models and it comparison with experimental data of a near-spherical particle. The mass loss history predicted by this work and non-isothermal model is in good agreement with experimental data, while isothermal model underestimates the conversion at the beginning and overestimate the conversion at later time. Thus, isothermal model should be modified with the heat transfer corrected coefficients, H_T and H_R . As it can be seen in Fig. 5 (b), after modification, particle average temperature profiles predicted by the isothermal model is consistent with non-isothermal model, while the isothermal model overestimates the heating rate. A comparison of isothermal and non-isothermal model reported by Lu et al.[6] shown similar results. Therefore, the devolatilization of thermally-thick biomass particle are dominated by internal heat transfer.

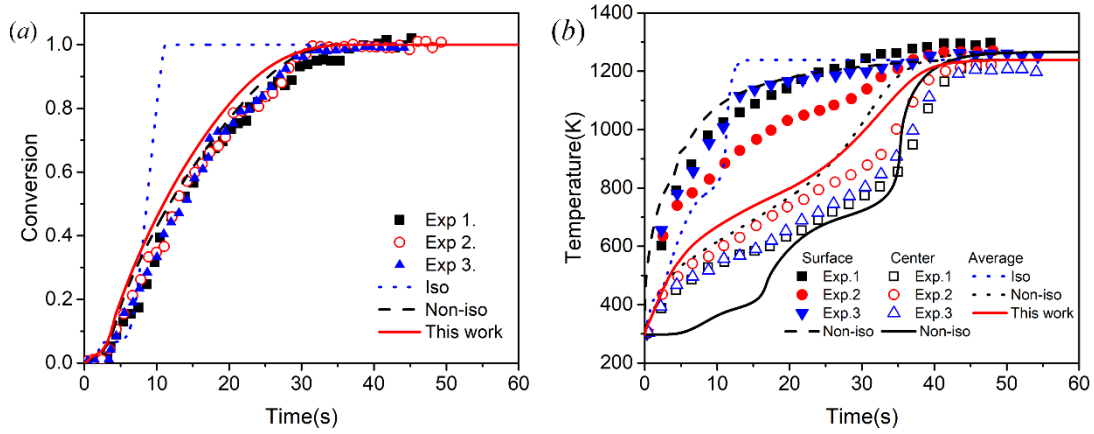


Figure 5. Total conversion (a) and temperature (b) comparison of near-spherical wood particle during devolatilization in nitrogen. (Conditions: $d_p=9.5$ mm, moisture content=6.38 dwt%, $T_g=1050$ K, $T_w=1276$ K)

For the cases of Alves and Figueiredo [10], the modeling of dry cylindrical wood particles are devolatilized at two different temperatures ($T_g=T_w=873$ K, and $T_g=T_w=1059$ K) are shown in Fig. 6. The results of these cases are similar with Fig. 5 (a). Isothermal model predicts lower conversion at the beginning time and high conversion at later time, and it underestimates the total conversion time as compared with experimental data. Moreover, when the isothermal model is used, a short flat conversion profiles are observed the modeling of wet cylinder wood particles, it means that drying happen first and then wood devolatilization occurs, it is not consistent with experimental data. While, the results predicted by non-isothermal model and this work are fitted well with experimental data for both dry and wet particle cases at different temperatures.

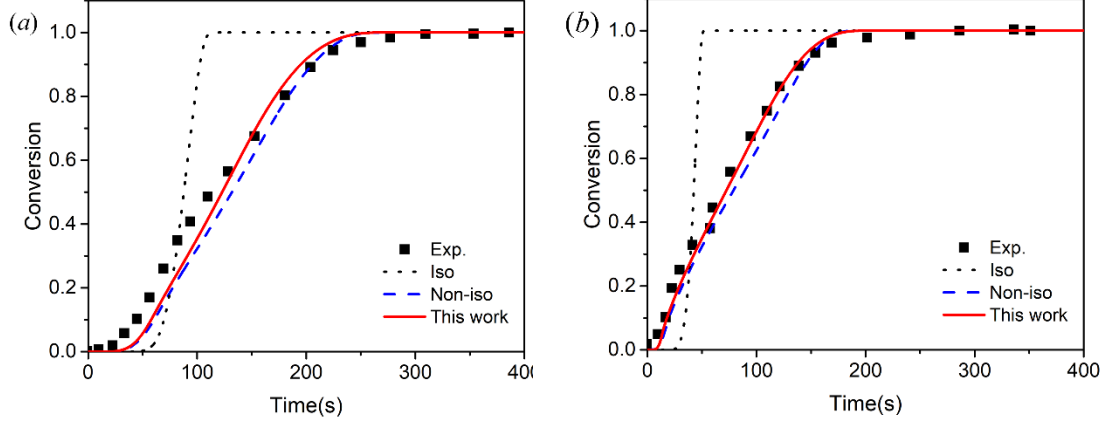


Figure 6. Total conversion comparison of dry wood particle during devolatilization in nitrogen at two different temperatures: (a) $d_p=18.5$ mm, $T_g=T_w=873$ K, (b) $d_p=19.4$ mm, $T_g=T_w=1059$ K.

4. Conclusions

In this paper, a heat transfer corrected isothermal model for biomass devolatilization by modifying heat transfer and reaction rates. Two heat-transfer corrected coefficients, H_T -correction of heat transfer and H_R -correction of reaction rates, are introduced. Those two coefficients are correlated as a binary function of heat convection coefficients and a dimensionless particle temperature. Thus, this model can be used to modeling both single biomass particle devolatilization and biomass devolatilization in fluidized bed. A series of single biomass devitalization case have been modeled to validate this work, the results predicted by this work are in well agreement with experimental data as well as consistent with the results of non-isothermal model for all size of particle, while isothermal model are only suit for small particle. Therefore, it is possible to apply this model to model devolatilization of large biomass particle without solving PDE equation in CFD simulation.

Notation

A_p	Particle surface area [m ²]	Bi	Biot Number [-]
C_p	Heat capacity [J/kg/k]	h_c	Heat convection coefficient [W/m ² /K]
H_R	Heat-transfer corrected coefficient for reaction rate	H_T	Heat-transfer corrected coefficient for heat convection
σ	StefaneBoltzmann constant [W/m ² /K ⁴]	ξ	Emissivity [-]
ρ_B	Density [kg/m ³]	m_p	Mass of particle [kg]
q_{iso}	Heat flux predicted by isothermal model [J/m ² /s]	$q_{non-iso}$	Heat flux predicted by non-isothermal model [J/m ² /s]
Q_R	Heat of reaction [J/kg]	R	Gas constant [J/mol/K]

R_{iso}	Reaction rate predicted by isothermal model [kg/s]	by $R_{non-iso}$	Reaction rate predicted by non-isothermal model [kg/s]
R_p	Particle reaction rate [kg/s]	t	Time[s]
T_g	Bulk gas temperature [K]	T_p	Particle temperature [K]
T_w	Wall temperature [K]	V_p	Particle Volume [m ³]
X	Conversion [-]	Y_v	Mass fraction of volatiles (Apparent fuel based) [-]
Y_{v0}	Initial mass fraction of volatiles in apparent fuel [-]	Y_w	Mass fraction of water (Apparent fuel based) [-]
Y_{w0}	Initial mass fraction of water in apparent fuel [-]		

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