



Modeling and simulation of combined pyrolysis and reduction zone for a downdraft biomass gasifier

Ningbo Gao, Aimin Li*

School of Environmental and Biological Science and Technology, Dalian University of Technology, Key Laboratory of Industrial Ecology and Environmental Engineering, MOE, Dalian 116024, PR China
School of Energy and Power Engineering, Dalian University of Technology, Dalian 116024, PR China

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ABSTRACT

This paper simulates the behavior of a global fixed bed biomass gasification reactor. The pyrolysis zone and reduction zone models are combined to simulate the global process of biomass gasification. The volatiles and gases released from the pyrolysis zone were assumed to crack into equivalent amounts of CO, CH₄ and H₂O. It is considered that the volatiles and gases leave the pyrolysis zone instantaneously and enter the reduction zone as initial gas concentrations. The numerical method applied is a Runge–Kutta fourth order method for solution of the pyrolysis zone model and finite differences for the reduction zone model to solve numerically the coupled ordinary differential equations. Simulations are performed for the varying pyrolysis temperature with a heating rate of 25 K/min and constant temperature of 1400 K as the initial reduction zone temperature at the same time. The simulation results for the temperature and concentrations of the gaseous species are in good agreement with published experimental data.

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1. Introduction

Biomass is one of the most promising renewable energy sources. Most biomass materials are available easily in wide areas. As it is abundant, environmentally friendly and renewable, the potential of biomass to help to meet the world energy demand has been widely recognized.

Thermochemical gasification is likely to be the most cost effective conversion process. Biomass gasification is one of the effective technologies for thermochemical conversion. The conversion is achieved by reactions between a feed gas and a feedstock. The primary goal of biomass gasification is optimal energy conversion of the solid biomass into a combustible gaseous product known as producer gas [1,2]. Three main thermal conversion layers are assumed in a gasifier, they are the combustion zone, pyrolysis zone and reduction zone. In an downdraft fixed bed, the biomass undergoes combustion, pyrolysis and gasification in turn. The oxygen is consumed at the grate; exothermic reactions mainly happen in this process, and the resulting hot gases (mainly including CO₂, N₂) cause char gasification and biomass pyrolysis. Understanding the chemical and physical mechanisms of the biomass gasification pro-

cess is essential to optimal designing and operating biomass gasification systems.

Biomass gasification is such a complicated process that it is difficult to simulate the total reaction. Separating the actual process into pyrolysis and reduction zones can simplify the model and improve the accuracy. Several authors have studied kinetic and equilibrium models of a downdraft biomass gasifier. Zainal et al. have studied an equilibrium gasification model based on equilibrium constants to simulation the gasification process in a downdraft gasifier [3]. Melgar et al. combine the chemical equilibrium and thermodynamic equilibrium of the global reaction to predict the final composition of the producer gas [4]. Giltrap and Babu present a model of the reduction zone of a downdraft biomass gasifier to predict the composition of the producer gas under steady state [5,6]. Srivastava et al. predict the concentration profiles in the cases of pyrolysis of different biomass materials in isothermal and non-isothermal conditions [7]. Koufopoulos et al. [8] presented a two step mechanism scheme for describing the kinetics of biomass pyrolysis, and Babu and Chaurasia [9] attempted to find the optimum kinetic modeling parameters for the pyrolysis process.

Giltrap and Babu's reduction zone model of biomass gasification only considered the fixed initial gas concentrations produced in the pyrolysis zone. However, the volatiles and gases of biomass pyrolysis vary instantaneously with different factors (e.g. time, temperature, heating rate etc.). Therefore, the volatiles and gases leave the pyrolysis zone and enter the reduction zone as initial gas concentrations that vary accordingly. Additionally, the initial

* Corresponding author. Address: School of Environmental and Biological Science and Technology, Dalian University of Technology, Key Laboratory of Industrial Ecology and Environmental Engineering, MOE, Dalian 116024, PR China. Tel./fax: +86 0411 8470 7448.

E-mail address: leeam@dlut.edu.cn (A. Li).

$$k_1 = A_1 \exp[(D_1/T) + (L_1/T^2)], \quad k_2 = A_2 \exp[(D_2/T) + (L_2/T^2)],$$

$$k_3 = A_3 \exp[(-E_3/RT)]$$

and

$$A_1 = 9.973 \times 10^{-5} \text{ s}^{-1}, \quad A_2 = 1.068 \times 10^{-3} \text{ s}^{-1},$$

$$A_3 = 5.7 \times 10^5 \text{ s}^{-1}, \quad D_1 = 17254.4 \text{ K},$$

$$D_2 = 10224.4 \text{ K}, \quad L_1 = -9061227 \text{ K}^2, \quad L_2 = -6123081 \text{ K}^2,$$

$$E_3 = 81,000 \text{ J/mol}$$

The initial conditions are as follows:

$$C_B = 1, \quad C_{G_1} = C_{C_1} = C_{G_2} = C_{C_2} = 0 \quad \text{at} \quad t = 0 \quad (6)$$

The temperature of the gasifier can be controlled, and a linear function exists between the temperature and time

$$T = \phi t + T_0 \quad (7)$$

By atomic balance, in a typical biomass (CH_{3.03}O_{1.17}), each mole of pyrolysis gas contains 0.341 mol CO, 0.277 mol CH₄ and 0.382 mol H₂O; we assume that all the volatiles can be transformed in the pyrolysis. According to the proportions of CO, CH₄ and H₂O contained in the pyrolysis, the final gas concentrations of the pyrolysis zone can be calculated, and they will be input in the simulation of the reduction zone as initial gas concentrations.

2.2. Model of reduction zone

Based on the Giltrap assumption, all the oxygen from the air inlet has been consumed with the reaction of C + O₂ = CO₂, and the pyrolysis products have been completely cracked into components of low molecular weight [5,6]. Plenty of char is formed in the combustion and pyrolysis zones and is assumed to exist in the reduction zone. Although Wang et al. and Giltrap [6,10] thought that the water–gas shift reaction (CO + H₂O ↔ CO₂ + H₂) had little effect on the global gasification, it was considered in the present study as a part of the entire reactions in the gasification system. Five reactions are considered in the reduction zone to predict the concentrations of six gaseous species (CH₄, H₂, CO, CO₂, N₂, H₂O):

- Reaction 1: Boudouard reaction: C + CO₂ ↔ 2CO
- Reaction 2: Water gas (primary) reaction: C + H₂O ↔ CO + H₂
- Reaction 3: Methanation reaction: C + 2H₂ ↔ CH₄
- Reaction 4: Steam reforming reaction: CH₄ + H₂O ↔ CO + 3H₂
- Reaction 5: Water gas shift reaction: CO + H₂O ↔ CO₂ + H₂

Among the six gaseous species, N₂ comes from the inlet air. It does not participate in any reactions; it only works to dilute the product gas. The model assumes a cylindrical gasifier bed of uniform cross section area A with negligible radial variation in the properties of both the bed and the gas. The mass and energy equations were set up as follows [5,6]:

Mass equation:

$$\frac{dn_x}{dz} = \frac{1}{v} \left(R_x - n_x \frac{dv}{dz} \right) \quad (8)$$

Energy equation:

$$\frac{dT}{dz} = \frac{1}{v \sum_x n_x C_x} \left(- \sum_x r_i \Delta H_i - v \frac{dP}{dz} - P \frac{dv}{dz} - \sum_x R_x C_x T \right) \quad (9)$$

$$\frac{dv}{dz} = \frac{1}{\sum_x n_x C_x + nR} \left[\frac{\sum_x n_x C_x \sum_x R_x}{n} - \frac{\sum_i r_i \Delta H_i}{T} - \frac{dP}{dz} \left(\frac{v}{T} + \frac{v \sum_x n_x C_x}{P} \right) - \sum_x R_x C_x \right] \quad (10)$$

$$\frac{dP}{dz} = 1183 \left(\rho_{\text{gas}} \frac{v^2}{\rho_{\text{air}}} \right) + 388.19v - 79.896 \quad (11)$$

Five reaction rates are considered to have an Arrhenius type temperature dependence and to be proportional to the difference between the actual reactant/product ratio and the corresponding equilibrium ratio. So, the five reaction rates can be obtained as follows:

$$r_1 = nA_1 \exp \left(\frac{-E_1}{RT} \right) \cdot \left(P_{\text{CO}_2} - \frac{P_{\text{CO}}^2}{K_1} \right) \quad (12)$$

$$r_2 = nA_2 \exp \left(\frac{-E_2}{RT} \right) \cdot \left(P_{\text{H}_2\text{O}} - \frac{P_{\text{CO}} \cdot P_{\text{H}_2}}{K_2} \right) \quad (13)$$

$$r_3 = nA_3 \exp \left(\frac{-E_3}{RT} \right) \cdot \left(P_{\text{H}_2}^2 - \frac{P_{\text{CH}_4}^2}{K_3} \right) \quad (14)$$

$$r_4 = nA_4 \exp \left(\frac{-E_4}{RT} \right) \cdot \left(P_{\text{CH}_4} \cdot P_{\text{H}_2\text{O}} - \frac{P_{\text{CO}} \cdot P_{\text{H}_2}^3}{K_4} \right) \quad (15)$$

$$r_5 = nA_5 \exp \left(\frac{-E_5}{RT} \right) \cdot \left(P_{\text{CO}_2} \cdot P_{\text{H}_2} - \frac{P_{\text{CO}} \cdot P_{\text{H}_2\text{O}}}{K_5} \right) \quad (16)$$

where

$$A_i = A'_i \cdot \text{CRF} \quad (17)$$

and A' is the frequency factor for reaction i, CRF is a factor presented by Giltrap and Babu [6,9]; it represents the relative reactivity of different char types. The first four reaction values of the activation energies and frequency factors are taken from Wang and Kinoshita [10] and the last reaction values are calculated by Eq. (18), see Table 1. The parameters of A_i and E_i for Reaction 5 are calculated as follows:

$$\ln k_i = \ln A_i - E_i/RT \quad (18)$$

The equilibrium is a function of temperature T and is written as follows: $k = \exp(-\Delta G^0/RT)$, After considering the equation, $\frac{\Delta G^0}{RT} = -\ln k$ can be written as $\frac{d \ln k}{dT} = \frac{\Delta H^0}{RT^2}$, since ΔH⁰ is a function of temperature T, then,

$$\ln k = \ln k^0 + \frac{\Delta A}{R} \ln \frac{T}{T_0} + \frac{\Delta B}{2R} (T - T_0) + \frac{\Delta C}{6R} (T^2 - T_0^2) + \frac{\Delta D}{12R} (T^3 - T_0^3) \quad (19)$$

where $\ln k_0 = \frac{-\Delta G^0}{RT_0}$ and ΔA, ΔB, ΔC and ΔD are calculated for each equilibrium reaction, e.g. for the reaction from Reaction 1, C + CO₂ ↔ 2CO, Δ = 2CO – CO₂ – C.

The equations to determine the values of ΔA, ΔB, ΔC and ΔD can be written as

$$\Delta A = 2A_{\text{CO}} - A_{\text{CO}_2} - A_{\text{C}},$$

$$\Delta B = 2B_{\text{CO}} - B_{\text{CO}_2} - B_{\text{C}},$$

$$\Delta C = 2C_{\text{CO}} - C_{\text{CO}_2} - C_{\text{C}},$$

$$\Delta D = 2D_{\text{CO}} - D_{\text{CO}_2} - D_{\text{C}}.$$

where A, B, C and D is reported in the thermodynamic data tables-Chemical Properties Handbook-for the reported range [3,11].

Table 1
Frequency factor and activation energies values [10]

Reaction i	A _i (1/s)	E _i (kJ/mol)
1	3.616 × 10 ¹	77.39
2	15170 × 10 ⁴	121.62
3	4.189 × 10 ⁻³	19.21
4	7.301 × 10 ⁻²	36.15
5	2.824 × 10 ⁻²	32.84

3. Numerical solution and simulation

3.1. Initial conditions of pyrolysis model

Solutions of the models of the pyrolysis zone and the reduction zone are different. The former is solved with the fourth order Runge–Kutta method, and the models of the reduction zone are solved numerically by a finite difference method.

In the present simulation, different initial non-isothermal processes, with initial temperature 773 K, heating rate 25 K/min and constant temperature 1400 K, have been considered. As the pyrolysis process is very slow below a temperature of 773 K [9] and the reduction reaction is slow accordingly, the lower values of initial temperature are not considered in the present study. The initial conditions are used for solving the coupled ordinary differential Eqs. (1)–(6) as follows:

$$t = 0, \quad C_B = 1.0, \quad C_{G_1} = C_{C_1} = C_{C_2} = C_{C_3} = 0$$

When C_B (concentration of initial biomass) < 0.03 , pyrolysis is found to be very slow and of little practical importance [9], so it can be seen as the final concentration.

The reduction zone model, Eqs. (8)–(11), contain nine first order differential equations in the system variables n_x (where x denotes the different gas species considered), P , v and T that is solved by an explicit finite difference method.

3.2. Initial conditions of reduction model

According to Giltrap and Babu [5,6], a variable 'pyrolysis fraction' (f_p) was introduced to express the degree of pyrolysis process. In the present study, we take $f_p = 0.5$ to simulate the biomass cracking reaction. Air was employed as the gasification medium; it is assumed that the oxygen mixed in air was consumed by the combustion reaction with char entirely in the combustion zone. Carbon dioxide, as the combustion production gas, participates in the reactions in the reduction zone, and nitrogen, as an inert gas, remains in the producer gas. For the typical biomass, with empirical formula $CH_{3.03}O_{1.17}$, we assume that the pyrolysis products were cracked into equivalent amounts of CO, CH_4 and H_2O , and thus, 0.341 mol CO, 0.277 mol CH_4 and 0.382 mol H_2O is contained in each mole of pyrolysis gas [6]. In the reduction zone, as the result of the tar of pyrolysis being cracked into light molecular gas, it is assumed that the volatiles of pyrolysis contain equivalent

amounts of pyrolysis gas. Two types of initial temperature reduction zone models are taken in the present work. One is taken by the changing the pyrolysis zone temperature, which is increased with a heating rate of 25 K/min from an initial temperature of 773 K, and the other is taken at a constant value of 1400 K. The initial velocity is taken 0.699 m/s, which is reported by Senelwa [12]. The increasing of operation pressure is caused by entrained dust that enters with the product gases and pipe resistance. To overcome the pressure drop, the operating pressure should be above atmospheric pressure; 1.005 atm was taken to simulate the reduction zone model [5].

The parameter of C_{RF} denotes the reactive degree of different char types. Giltrap et al. found that the order of 1000 for C_{RF} produced a similar result with Senelwa's research. In this paper, the value of 1000 is also taken in the model.

4. Results and discussion

4.1. Validation of the model

Equilibrium constants of the reduction zone were compared with data collected from different sources as shown in Fig. 3. The equilibrium constants of the steam reformation reaction, Boudourd reaction and water–gas shift reaction increase with reaction temperature increasing, however, the methanation reaction and the water–gas shift reaction show inverse trends, the equilibrium constants decrease with temperature increasing. The predictions of equilibrium constants agree well with the data from the calculated JANAF tables.

The model developed above has been used to predict the gas composition, reaction temperature, unconverted char and calorific value of gas for known biomass composition, initial temperature of pyrolysis zone, velocity of air flow and pressure. Table 2 shows the parameters used in the comparison.

The simulation results concerning the model in the pyrolysis and reduction zones are in good agreement with different researchers' studies. Fig. 4 shows the concentration of materials produced in the pyrolysis zone with $n_1 = 1$ and $n_2 = n_3 = 1.5$ for the heating rate of 25 K/min. From the trend shown in the figure, it is similar to the results reported by Srivastava et al. [8]. Fig. 5 shows the predicted concentration of composition gas produced in constant temperature reduction zone. It is shown that the simulated concentrations of producer gas are in good agreement with the experimental data.

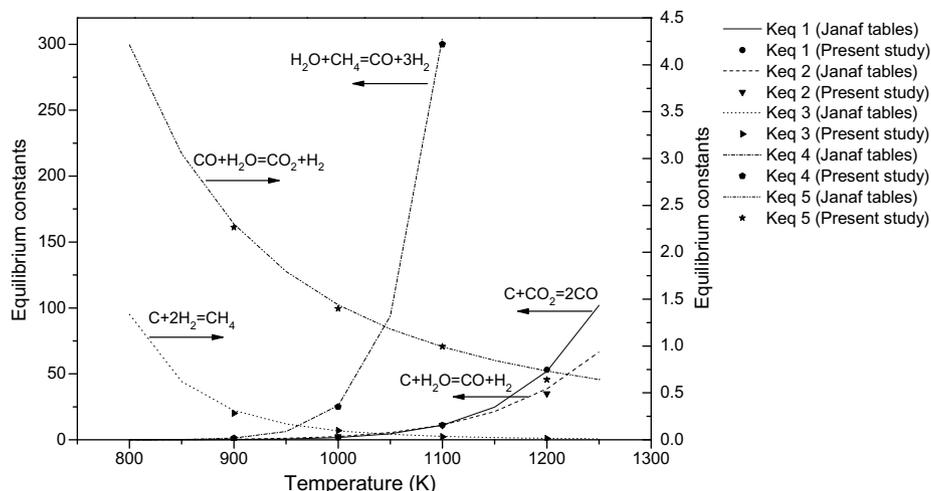


Fig. 3. Comparison of equilibrium constants predicted of reduction zone reactions with JANAF tables.

Table 2
Parameters used in experimental downdraft biomass gasification and in the model simulation

Parameter	Chee [13] data	Senelwa [12] data	Giltrap et al. [6] data	Present model
Bed length	Not stated	0.275 m	0.275 m	0.275 m
T_{initial}	Not stated	1228 K	1200 K	Initial value from pyrolysis zone
v	Not stated	0.699 m/s	1 m/s	0.699 m/s
Biomass	Cotton wood chip		Douglas fir bark	Douglas fir bark
C_{RF}			1000	1000
f_{D}			0.5	0.5
Moisture content	5 wt%	'Oven dried'	0	0

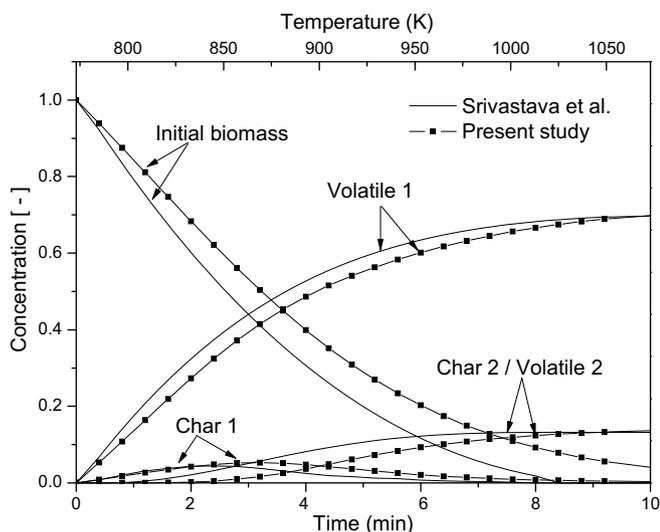


Fig. 4. Concentration of pyrolysis zone predicted by model compared with data reported in the literature.

4.2. Effect of heating rate of 25 K/min in pyrolysis zone

Fig. 6 shows the simulated profiles of temperature and concentrations of producer gas (CH_4 , H_2 , CO , CO_2 , N_2 , H_2O) in the reduction zone as the pyrolysis zone temperature increases with a heating rate of 25 K/min. Owing to the temperature increasing linearly, the initial biomass, volatiles and chars vary with time, and then, the concentrations of the gasification gases change also. The distribution of the concentrations of the different chemical species in

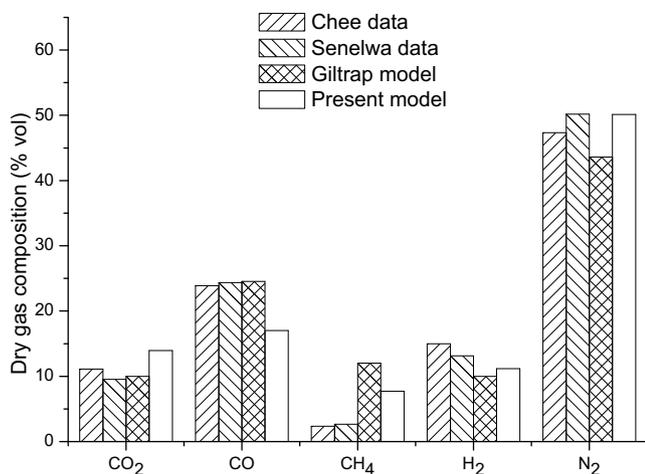


Fig. 5. Comparison of product gas predicted and experimental results.

the producer gas composition depends mainly on the chemical equilibrium between the species, and temperature is a key factor in all reactions. As shown in Fig. 6f, the greatest temperature gradient takes place in the first 30 mm from the top of the reduction zone in the whole reaction time, and in the next 245 mm, the temperature variation is less and more smooth. At the section of 30 mm distance downstream of the reduction zone, strong reactions (Reactions 1–5) occurred. Although the transported temperature from the pyrolysis zone increased linearly, the temperature along the reduction zone decreased rapidly. As a result, the concentrations of the gasification gases in the same region have similar changes.

The variations of the concentrations of the gasification products in the reduction zone along the axis distance at the final time of gasification (heating rate is 25 K/min in pyrolysis zone) was shown in Fig. 7. The concentrations of hydrogen and carbon monoxide are increasing along the length of the reduction zone with the increasing reaction time, however, the concentrations of methane, nitrogen, carbon dioxide and water content are decreasing. Although the formation of methane during the gasifying process depends directly on the temperature reached in the reaction [4], in the effect of steam reforming reaction (Reaction 4), the concentration of methane is decreasing. Because of Reaction 1, carbon dioxide was transformed into carbon monoxide, increasing the concentration of carbon monoxide. Although nitrogen, as an inert gas, is not participating in any reaction and its amount remains steady, the amounts of the other species of gas are changing. So, their concentrations vary accordingly. Because of the high heating value of hydrogen, the increasing hydrogen fraction improved the quality of the producer gas composition. Although nitrogen is inert, it may affect the reactions in biomass gasification in three respects [10]. The first one is in diluting the reactants concentration; as a result, the reaction rates are decreased. The second is reducing the residence time; longer residence time makes reaching chemical equilibrium more easy, and higher heating value producer gas is produced. Finally, nitrogen may inhibit char reactions because its molecules cover some active char surface.

Fig. 8 shows the concentrations of the gasification products at the bottom (at 0.275 m) of the reduction zone with reaction time. The concentrations of hydrogen and carbon monoxide increase while those of nitrogen and carbon dioxide decrease with increasing reaction temperature. The methane content undergoes a process of increasing from 0% to 11.16% at the eighth minute, and at the end of the reaction, the concentration decreases to 10.83%. Similarly, the water content reached the maximum fraction of 15.23% at 7.8 min and finally decreased to 14.60%. As shown in Fig. 8, the temperature of this point is increasing from 732 K to 882 K.

4.3. Effect of constant temperature 1400 K in pyrolysis zone

The effect of constant temperature in the pyrolysis zone was studied in the present study. Fig. 9 shows the composition profile

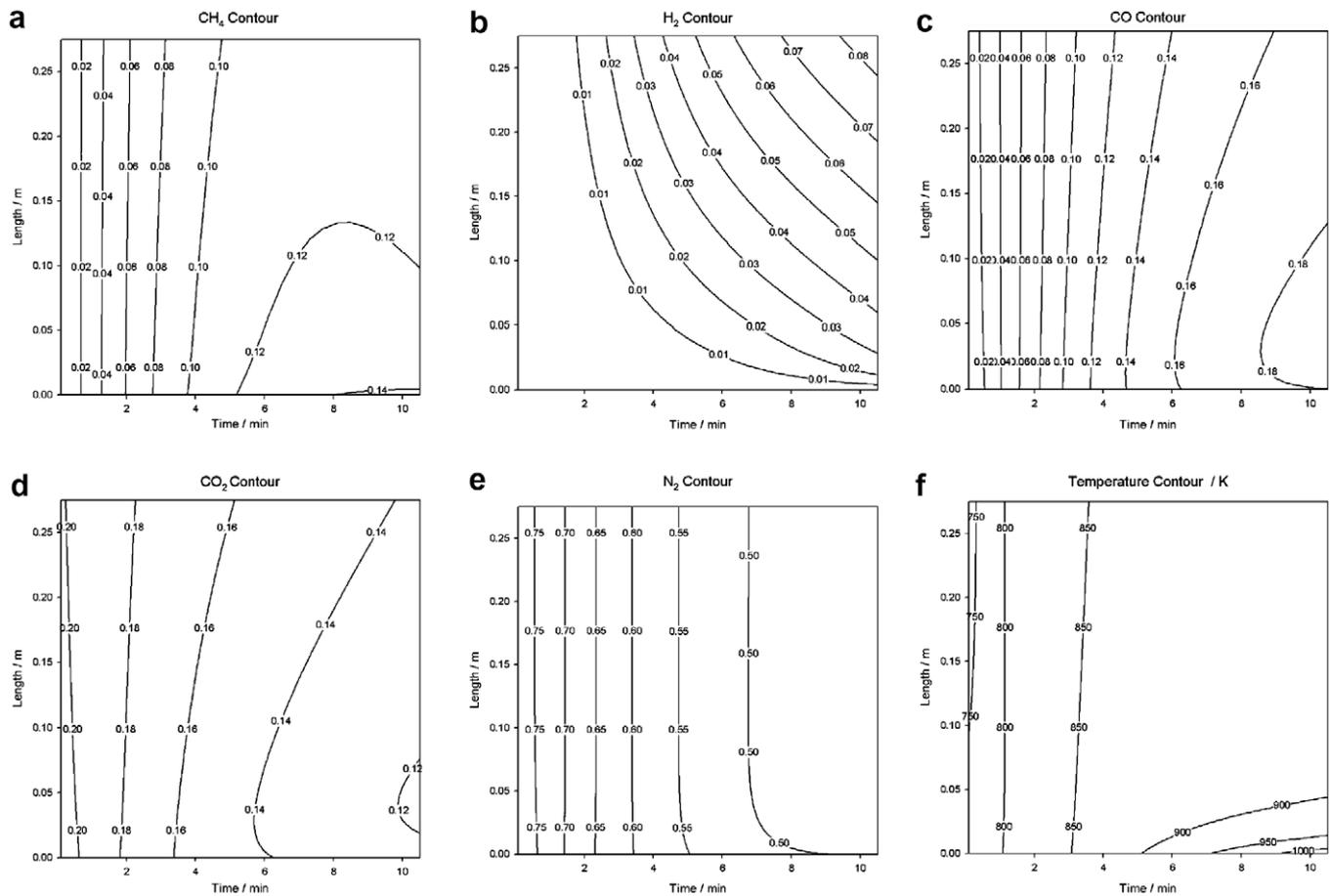


Fig. 6. Simulated profiles of producer gas concentration and temperature of reduction zone as the pyrolysis zone temperature increasing with heating rate of 25 K/min.

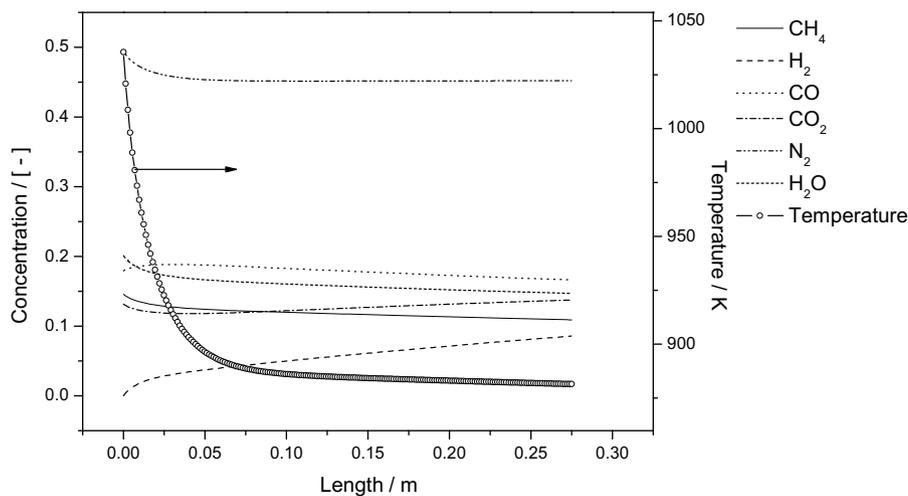


Fig. 7. Species concentration of gasification products along reduction zone length at final time of gasification.

at the end of the reduction zone for the constant temperature of 1400 K in the pyrolysis zone. The temperature increased from 884 K to 932 K, and in the first 3 min, the temperature gradient is obviously higher than that later. Nitrogen and carbon dioxide decrease and methane, hydrogen, carbon monoxide and water content increase with reaction time.

Fig. 10 shows the concentration profiles at different lengths of the reduction zone at the final gasification time, 12.2 min. During

the first 61 mm length of the reduction zone, the temperatures decrease rapidly from 1400 K to 940 K. The chemical reactions proceed very rapidly during this zone. The reason for this is that in the reduction zone, all reactions except Reactions 3 and 5 are endothermic, and the heat release from Reactions 3 and 5 is less than that of the other reactions. As a result, the system temperature decreases rapidly. The concentrations of all species vary acutely also. From 61 to 275 mm, the temperature varies relatively slowly. Sim-

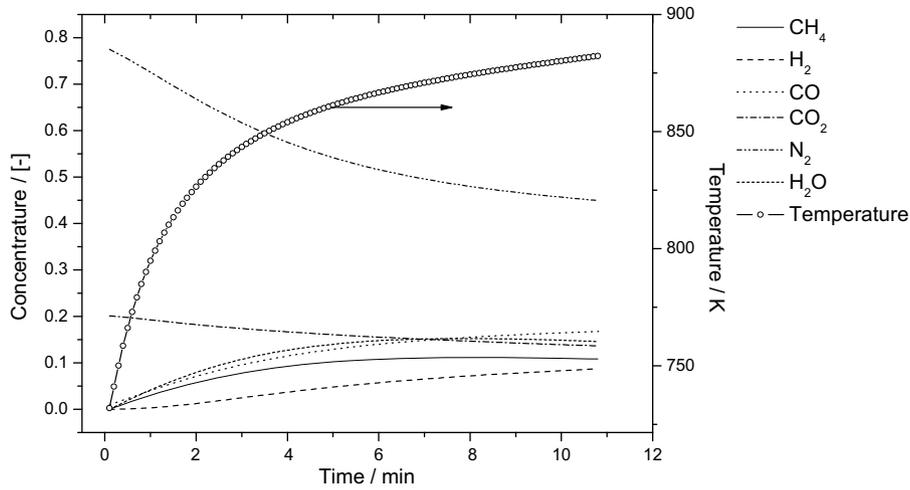


Fig. 8. Composition profile at the bottom of reduction zone for the heating rate of 25 K/min.

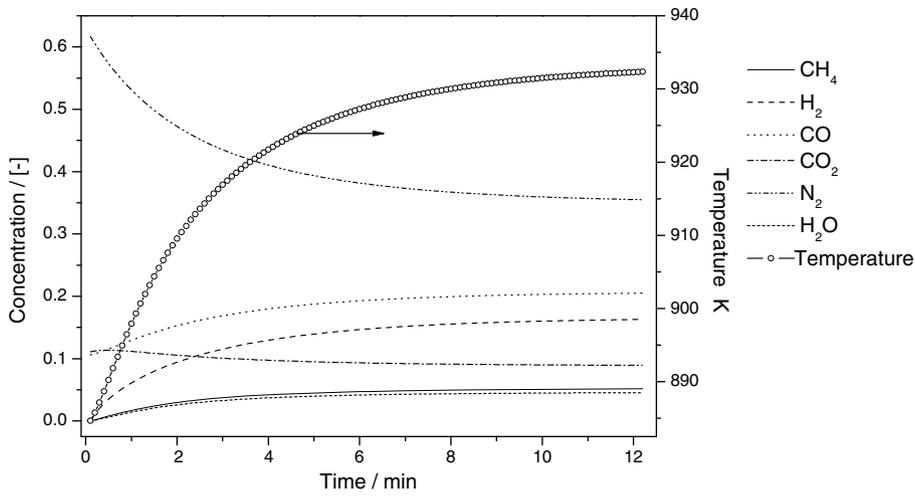


Fig. 9. Composition profile at the end of reduction zone for the constant temperature of 1400 K in pyrolysis zone.

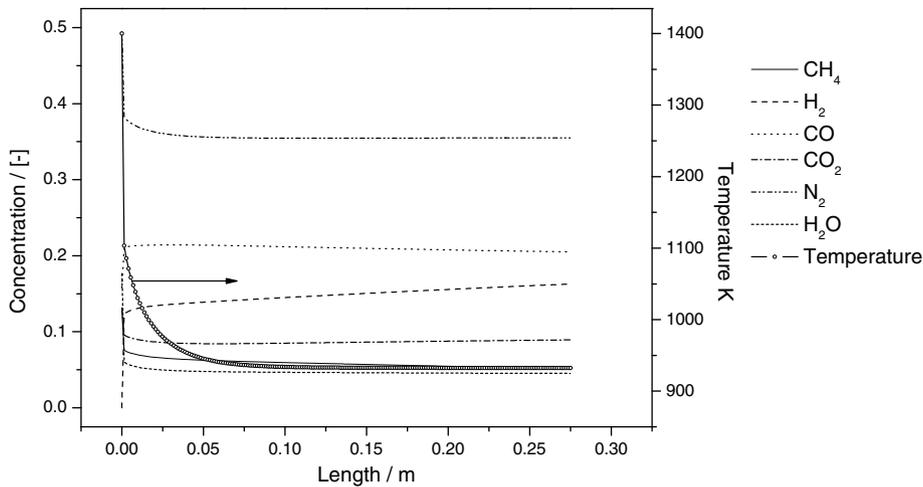


Fig. 10. The final composition profile in reduction zone for constant temperature 1400 K at pyrolysis zone.

ilarly, the chemical reactions process shows flat trends. This may be caused by the char surface area decreasing due to carbon consumption, and the produced hydrogen and carbon monoxide pre-

vent further reactions from the covered carbon surface [10]. Fig. 11 shows the temperature profile in the whole reaction field. The trend is that the temperature has a sharp variation at the inlet

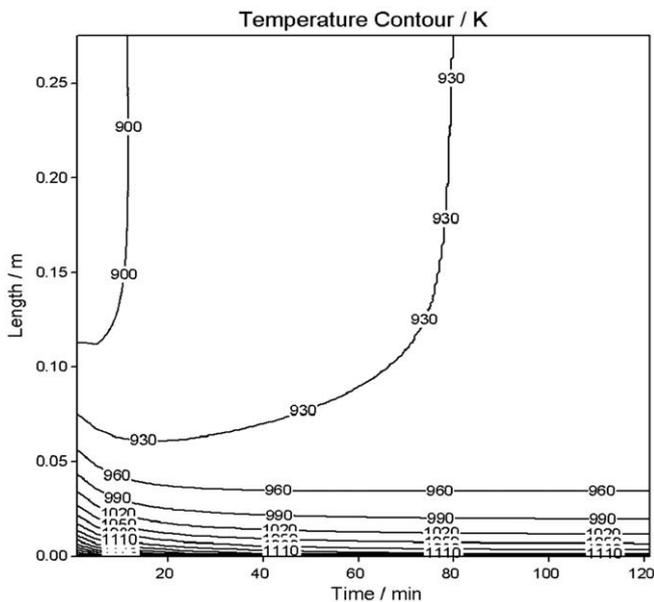


Fig. 11. The profile of reduction zone temperature for constant temperature 1400 K at pyrolysis zone.

side of the reduction zone, and thereafter, the trend varies relatively slowly.

5. Conclusions

A combined pyrolysis zone and reduction zone model was used in the present study in order to simulate the biomass gasification process. The earlier model reported by Giltrap et al. and Babu researched the pyrolysis zone and reduction zone of gasification independently. The model considered the biomass gasification as

an integrated process and predicted the trends of temperature field and producer gas concentration field in time and space. Two modes are performed for varying the temperature of the pyrolysis zone, one is using a heating rate of 25 K/min to heat the pyrolysis zone, and the other is using a constant value of 1400 K as the pyrolysis temperature. In the two modes, the trends of temperature profile and species concentrations are very different.

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