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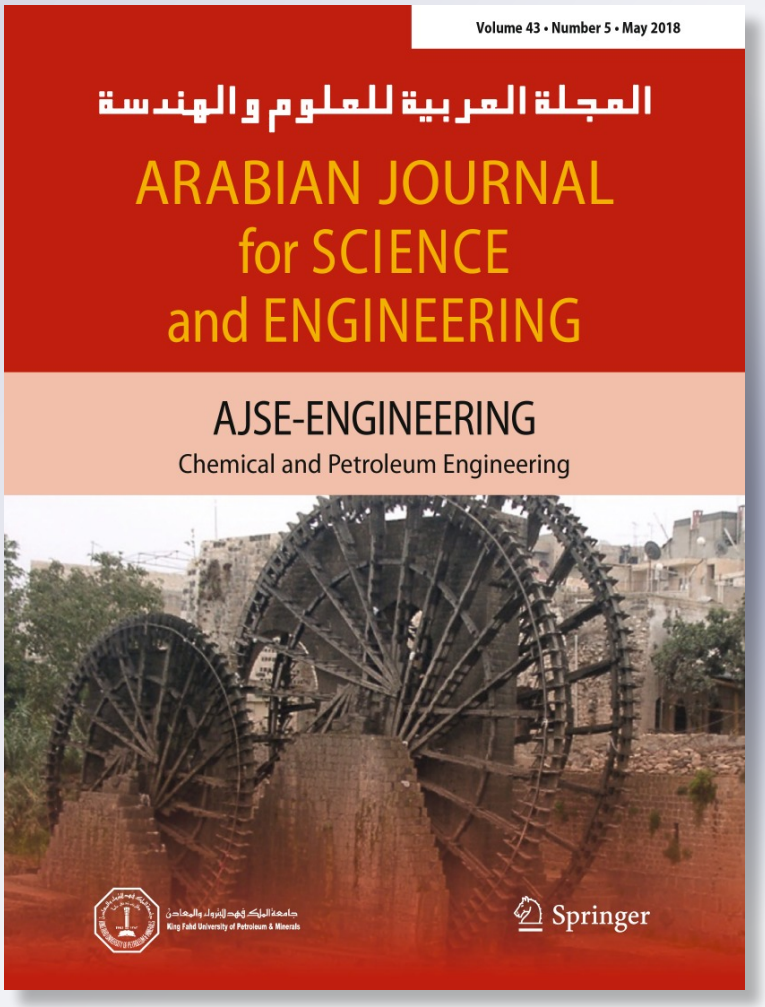

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RESEARCH ARTICLE - CHEMICAL ENGINEERING

Thin Reaction Zone Model for Evaluating the Mechanisms that Control the Char Gasification Process: 1. Quasi-steady State

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Abstract Reaction mechanism takes place in char gasification was tested by kinetic studies. A modified kinetic model (thin reaction zone model) was developed to evaluate the mechanisms controlling the process of gasification. The model was compared to the experimental data of char gasification using steam as gasification agent. The gasification was carried out at 600–800 °C for 60 min. The model assuming the mass transfer of steam through the ash layer controls the reaction rate work well. The average error at operating temperatures of 600, 700, and 800 °C was 2.89, 1.75, and 3.26% respectively.

Keywords Char gasification · Kinetic parameter · Mass transfer through ash layer · Thin reaction zone model

List of symbols

D_e	Molecular diffusivity ($cm^2\ min^{-1}$)
C_g	Concentration of gas A ($gmol\ L^{-1}$)
k_{TRZM}	TRZM rate constant (min^{-1})
k	Rate constant of the reaction (min^{-1})
M	Molecular weight ($g\ mol^{-1}$)
R	Initial radius of solid particle (cm)
X_B	Carbon conversion
ρ	Density of solid particle ($g\ L^{-1}$)
ε	Thickness of unreacted zone (cm)

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1 Introduction

Indonesia has enormous potential of lignite and subbituminous coal. Those types are categorized as low rank, so to optimize their utilization, it is necessary to upgrade the quality. One upgrading method to increase the coal quality as energy source is gasification. It is considered as an effective and efficient method for converting solid carbonaceous matter to the gas producer or synthetic gas. This gas can be used for power and heat generation or a synthesis precursor [1,2].

However, gasification is a complex process. Understanding the influence of raw material, operating conditions, and the interaction between the chemical and physical mechanisms during gasification is basically important to search for an optimal design of gasifier [3–6].

On commercial plant, the optimum operating conditions were often obtained through observation on the operating unit or through experiments on the pilot plant. In addition to the experimental observation, the optimum conditions of a gasification process can be also be obtained from mathematical modeling. Although it is only result of theoretical predictions, mathematical models can provide guidance regarding the influence of quantitative design, raw material, and other operating parameters. Moreover, the representation of the gasification process in the form of mathematical models can assist in gaining insights into the importance of the operating parameters affecting the performance of the gasifier [1,2,6].

The kinetic parameters are very important to get information about the reaction mechanism take place in the char gasification process [7–10,12]. To understand the kinetic parameters, a number of studies have been studied widely. Some researchers have developed kinetic models to analyze reaction kinetic in the process of gasification such as volumet-

ric model (VM), shrinking core model (SCM), random pore model (RPM), and active site intermediate model (ASIM).

Volumetric model assumes that during the process of char gasification, active sites are uniformly distributed on the surface of the char and particle size remains constant [11,12]. Nevertheless, for un-catalytic gasification process, this model can predict char conversion as a function of reaction time and is acceptable for the char conversion less than 75% [14].

Shrinking core model assumes that reaction occurs first at the outer skin of particle then moves into the solid [15]. Mass and porosity of char are distributed homogeneously, and surface area exposed to gasification agent is proportionate to the amount of the active sites of the char [14]. This model is widely used to describe the char gasification with shape factor $m = 2/3$ [14].

Nevertheless, both VM and SCM cannot describe catalyzed char gasification [14–16]. For this reason, this study will develop a kinetic model based on the mechanism of mass transfer and the surface reaction on the active sites of the char. It aims to know the mechanisms that control the process of gasification.

To determine the controlling mechanism and to calculate the parameter values of the mass transfer and reaction kinetics of the model, experiment was conducted on the char gasification using steam as gasification agent. The gasification was carried out in a updraft reactor at atmospheric pressure and at three different temperatures; i.e., 600, 700, and 800 °C for 10 g of char.

2 Model Development

In this study, a kinetics equation for shrinking core model is developed based on a model set up by Yagi and Kunii with a slight modification [15]. The modification is founded on the assumption that the diffusion of steam through the ash layer is in quasi-steady state.

It is considered that irreversible reaction occurs on the surface of spherical solid can be described as follow:

$$A_{(g)} + B_{(s)} \rightarrow P_{(g)} + Q_{(s)} \tag{1}$$

The mechanisms of the reaction are explained in Fig. 1: Assumptions used in model development were:

(a) The rate of mass transfer through the gas film around particle solid are relatively fast, so it does not control the overall processes.
(b) The size of the solid product does not change during the process.
(c) The size of unreacted zone changes at any time.

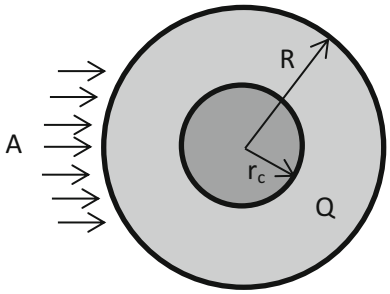


Fig. 1 Representation of mechanisms occurs in model development

(d) The reaction occurs at the surface of the unreacted zone at thickness εε, and the value of ε is constant and relatively thin.

Gas A diffuses into the unreacted zone through the solid product (Q). Mass balance of A in the solid product are follows Eq. (2):

$$\frac{\partial^2 C_A}{\partial r^2} + \frac{2}{r}\frac{\partial C_A}{\partial r} = -\frac{1}{D_e}\frac{\partial C_A}{\partial t} \tag{2}$$

If the diffusion is considered quasi-steady state, Eq. (2) can be expressed in the following form:

$$\frac{d^2 C_A}{dr^2} + \frac{2}{r}\frac{dC_A}{dr} = 0 \tag{3}$$

Boundary conditions

1. Based on the assumption a, the concentration of gas A at the solid surface is equal to the concentration of gas A in the bulk of gas. So the first boundary condition can be written as:

$$C_A = C_g \text{ at } r = R \tag{4a}$$

2. The concentration of gas A at the solid surface (C_{Ai}) of the unreacted zone (r_c) changes at any time. Let the concentration be C_{Ai}, so:

$$C_A = C_{Ai} \text{ at } r = r_c \tag{4b}$$

Equation (3) with the boundary conditions of Eqs. (4a) and (4b) can be solved analytically and the result is:

$$C_A = \frac{1}{1 - r_c/R}\left[C_g - (r_c/R)\,C_{Ai} - \frac{r_c}{r}\left(C_g - C_{Ai}\right)\right] \tag{5}$$

where C_A is a concentration of gas A at any position of r in the solid product.

Determination of C_{Ai}

The equation of C_{Ai} can be evaluated by setting the material balance of A at the reaction zone on the surface of the unreacted core as follows:

$$\begin{pmatrix}\text{The rate of}\\ \text{accumulation}\\ \text{of } A\end{pmatrix} = \begin{pmatrix}\text{The rate of}\\ \text{diffusion}\\ \text{of } A \text{ at } r = r_c\end{pmatrix} - \begin{pmatrix}\text{The rate of}\\ \text{reaction of } A\\ \text{at the reaction zone}\end{pmatrix}$$

$$4\pi r_c^2 \varepsilon \frac{dC_{Ai}}{dt} = 4\pi r_c^2 D_e \left.\frac{dC_A}{dr}\right|_{r=r_c} - 4\pi r_c^2 \varepsilon k C_{Ai} \tag{6}$$

If C_{Ai} is assumed to be quasi-steady state $\left(\frac{dC_{Ai}}{dt} = 0\right)$, it can be obtained that:

$$C_{Ai} = \frac{D'_e}{k} \left.\frac{dC_A}{dr}\right|_{r=r_c} \tag{7}$$

where $D'_e = \frac{D_e}{\varepsilon}$

Differentiation of Eq. (5) gives:

$$\left.\frac{dC_A}{dr}\right|_{r=r_c} = \frac{(C_g - C_{Ai})}{r_c\,(1 - r_c/R)} \tag{8}$$

Substitution of Eq. (8) into Eq. (7) will result in:

$$C_{Ai} = \frac{D'_e}{(1 - r_c/R)} \frac{C_g}{\left[\frac{D'_e}{(1-r_c/R)} + k r_c\right]} \tag{9}$$

where r_c are unreacted zone radius which changes at any time.

Determination of r_c

The rate of depletion of B is equal to the rate of reaction, so:

$$\frac{d}{dt}\left(\frac{4}{3}\pi r_c^3 \rho \frac{1}{M}\right) = -4\pi r_c^2 \varepsilon k C_{Ai} \tag{10}$$

Equation (10) can be expressed in the following form:

$$\frac{dr_c}{dt} = -\frac{M\varepsilon}{\rho} k C_{Ai} \tag{11}$$

Substitution Eq. (9) into Eq. (11) will result in Eq. (12):

$$\left(1 - \frac{r_c}{R}\right) + \left(\frac{R}{D'_e}\right) k \left[\frac{1}{2}\left(1 - \left(\frac{r_c}{R}\right)^2\right) - \frac{1}{3}\left(1 - \left(\frac{r_c}{R}\right)^3\right)\right] = \left(\frac{M\varepsilon C_g}{R\rho} k\right) t \tag{12}$$

Equation (12) can be written in terms of conversion by considering that:

$$X_B = \frac{4/3\pi R^3 - 4/3\pi r_c^3}{4/3\pi R^3} = 1 - \left(\frac{r_c}{R}\right)^3 \tag{13}$$

Therefore, Eq. (12) can be written in terms of conversion as follows:

$$\left(\frac{1}{k_{TRZ}}\right)\left[1 - (1 - X_B)^{\frac{1}{3}}\right] + \left(\frac{1}{\alpha}\right)\left[\frac{1}{2} - \frac{1}{2}(1 - X_B)^{\frac{2}{3}} - \frac{1}{3}X_B\right] = t \tag{14}$$

where

$$k_{TRZ} = \left(\frac{M\varepsilon C_g}{R\rho}\right) k \tag{15a}$$

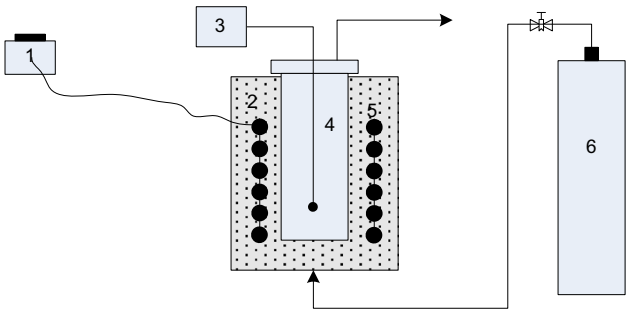
$$\alpha = \left(\frac{MC_g}{R^2\rho}\right) D_e \tag{15b}$$

3 Experimental Setup

3.1 Preparation of Char

Pattukku coal obtained from Lappariaja area, district of Bone, South Sulawesi was ground and sieved to a particle size of 3.35–4 mm. The pyrolysis was carried out in a quartz tubular reactor with a diameter of 3.5 cm and a height of 55 cm at 450 °C and 1 atm. A schematic diagram of the pyrolysis equipment is shown in Fig. 2.

One hundred grams of coal was fed into the reactor. Nitrogen was flowed from the bottom of the reactor for 15 min to eliminate the existing air in the reactor. Then pyrolysis was started by switching on the electric furnace to heat the reactor. The whole experiment was carried out at fixed temperature



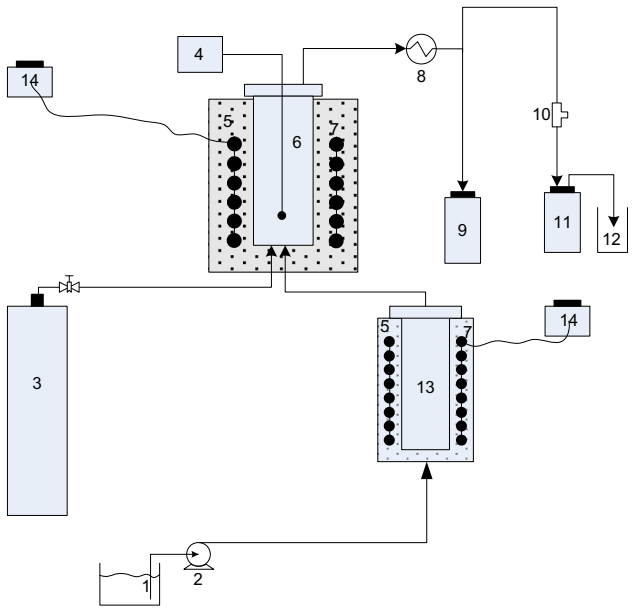
1. Regulator
2. Isolator
3. Thermocoupel
4. Reactor
5. Coil heater
6. Nitrogen tank

Fig. 2 The schematic diagram of the pyrolysis apparatus

of 450 °C with heating rate at 30 °C/min. When the temperature reached 450 °C, the controller was turned on and the isothermal process was held for 60 min. After that, the reactor was left until the temperature reached 30 °C.

3.2 Gasification Process

The schematic diagram of the gasification equipment is presented in Fig. 3. Ten grams of char was fed into the gasifier during each experiment. Before the process started, nitrogen was flowed from the bottom of the reactor to remove air from the reactor. The experiment was started by switching on the electric furnace to increase the temperature of the gasifier. After the desired temperature was reached, steam was flowed from the bottom of the reactor at the rate of 5 mL/min. The non-condensed gas product was collected continuously in a tank containing water after it passed through the condenser. The volume of the collected gas was measured at the various time. Gasification process was carried out for 60 min, in which every 5 min, samples were taken for analysis. The concentration of CH_4, CO, H_2 and CO_2 was measured with gas chromatography. The procedure was repeated for temperature 600, 700, and 800 °C.



1. Water tank
2. Pump
3. Nitrogen tank
4. Thermocoupel
5. Coil heater
6. Reactor
7. Isolator
8. Condenser
9. Condensate tank
10. Sampling gas
11. Water tank
12. Water reservoir
13. Steamer
14. Regulator

Fig. 3 The schematic diagram of the experimental apparatus

4 Result and Discussion

4.1 Char Conversion

In this study, steam gasification is running 60 min in three different temperatures; i.e. 600, 700, and 800 °C. The carbon conversions for each temperature are presented in Fig. 4.

Figure 4 shows the carbon conversion varies with time in the steam gasification of char at 600, 700, and 800 °C for 60 min. Carbon conversion was calculated based on mols of CO, CO_2, and CH_4 by using Eq. (16) as follows

$$X_B = \frac{n_{CO} + n_{CO_2} + n_{CH_4}}{n_{C_0}} \times 100\% \tag{16}$$

From Fig. 4 it can be seen that in general, the carbon conversion will increase by time significantly until 30 min and then it goes relatively slowly. The final carbon conversion at gasification temperature of 600, 700, and 800 °C are 2.02, 2.95, and 4.43% respectively.

This indicates that temperature is one of the factors that can increase conversion reaction. In higher temperature, the kinetic energy of molecules will also be greater so that collisions among molecules become more frequent and this causes the reaction occurs faster.

4.2 Validation of the Model and Evaluation of Parameters

To evaluate the value of each parameter in Eq. (14), the curve fitting method is used in which the value of Sum of Square of Error (SSE) is minimized.

$$\mathrm{SSE} = \sum_{i=1}^{m} \left(\frac{X_{data} - X_{Calculate}}{X_{data}} \right)^2 \tag{17}$$

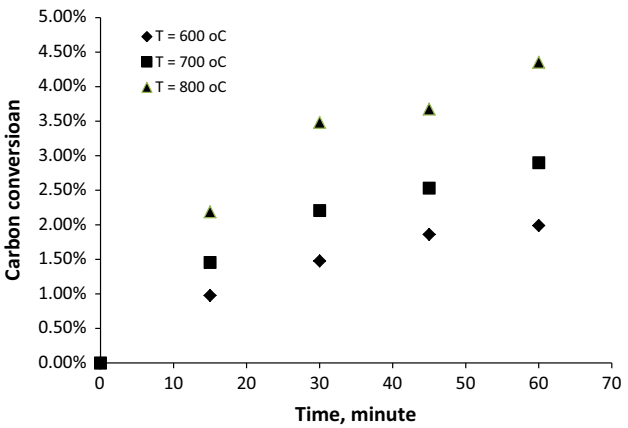


Fig. 4 The relationship between reaction time and carbon conversion at steam gasification temperature 600, 700, and 800 °C for 60 min

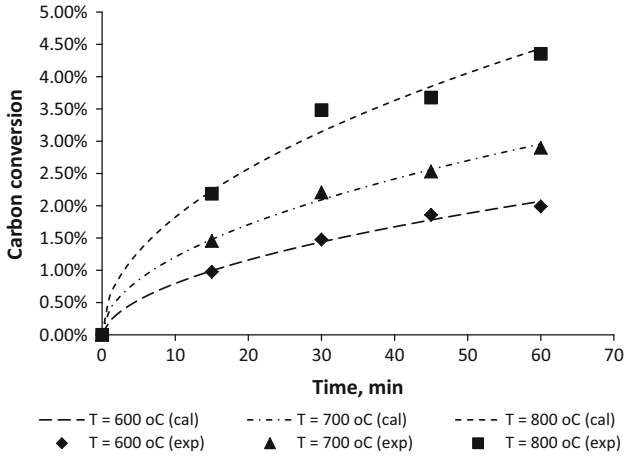


Fig. 5 Comparison of experimental data and calculated char conversion at steam gasification temperature 600, 700, and 800 °C for 60 min using thin reaction zone model

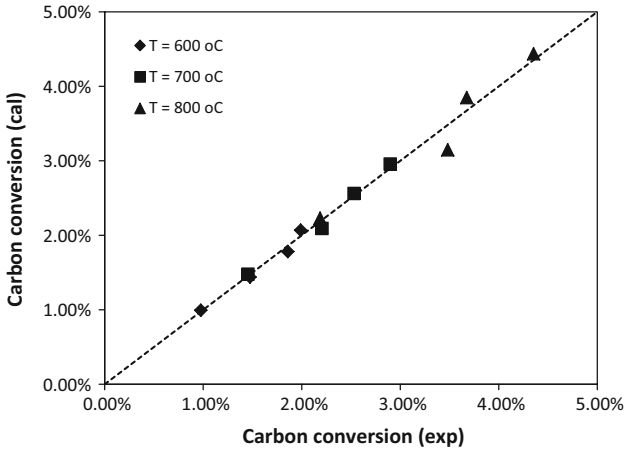


Fig. 6 The deviation of the calculated carbon conversion toward the result of experimental at temperature 600, 700, and 800 °C for 60 min using thin reaction zone model

The comparison of experimental data and the calculated result for each temperature can be seen in Fig. 5.

Figure 5 shows the relationship between reaction time and char conversion in steam gasification at 600–800 °C for 60 min. The char conversion is defined as the total moles of CO, CO_2, and CH_4 divided by total moles of carbon in the char that fed into the gasifier [Eq. (16)].

As shown in Fig. 5, the char conversion from the calculation result using the thin reaction zone model fits well with the result obtained from the experimental data. The deviations of the results of thin reaction zone model to experimental data for all temperatures are presented in Fig. 6. It can be visually observed that the thin reaction zone model was matched to experimental data.

From the result of the calculation, it is found the average the deviation at 600, 700, and 800 °C were 2.5, 2.00, and 3.64%, respectively. These indicated that thin reaction zone model is able to illustrate the process mechanisms properly of char gasification.

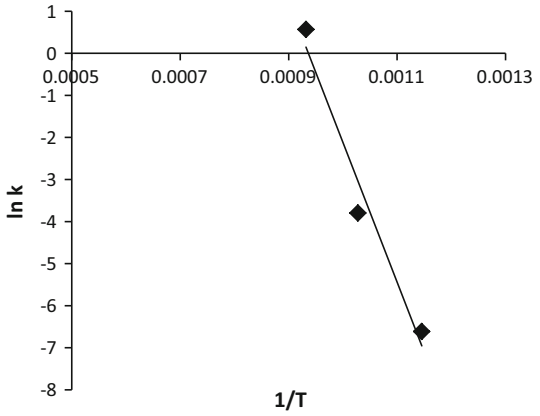


Fig. 7 The Arrhenius's plot of ln$k$ versus $1/T$ for a range temperature of gasifier 600–800 °C

(a) Evaluation of the kinetic parameters (k_{TRZM})

Activation energy (E_a) and frequency factor (k_o) can be obtained by plotting ln$k$ versus $1/T$ as shown in Fig. 7. This plot was obtained by calculation of reaction rate constant at three different temperatures.

Based on Fig. 7, the value of the kinetic parameters (E_a and k_o) were 98.197 kJ/mol and 3.129×10^4 min^{-1}, respectively. Thus, the equation of the reaction rate constant can be written as follows:

$$k_{TRZM} = 3.129 \times 10^4 e^{\frac{-98.197}{RT}} \quad (18)$$

These results are compared with other investigators as presented in Table 1.

(b) Evaluation of the mass transfer parameter (α)

Based on the simulated result, the relation between mass transfer parameter (α) and temperature (T) is shown in Fig. 8.

From Fig. 8, the equation of α as a function of temperature is as follows:

$$\alpha = \alpha_o e^{\left(\frac{-\alpha_1}{T}\right)} \quad (19)$$

Furthermore, Eq. (19) can be written in a linear equation as follows:

$$\ln \alpha = \ln \alpha_o - \frac{\alpha_1}{T} \quad (20)$$

Linear regression was used to determine the constants of α_o and α_1 and the results are 1.983×10^{-3} min^{-1} and 7439.7

Table 1 Comparisons of kinetic parameters with other investigators

Investigator	Temperature (°C)	E_a (kJ/mol)	Pre-exponential factor, min^{-1}
Ye et al. [13]	714–892	131	2.61×10^5
Tang et al. [14]	750–950	91.57	1.12×10^3
Karimi et al. [11]	600–800	210	3.78×10^5
This research	600–800	98.197	3.129×10^4

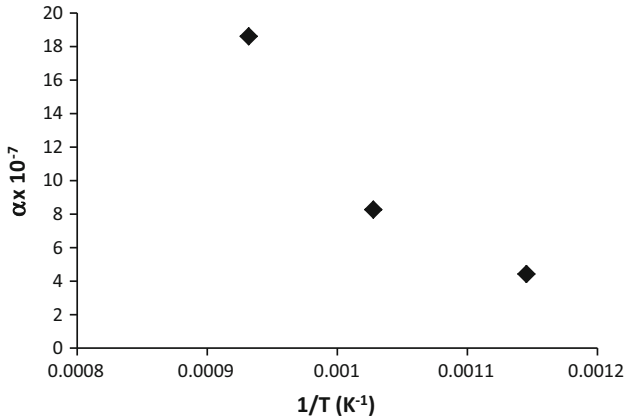


Fig. 8 The mass transfer parameter (α) as a function of $1/T$

K respectively. Thus, the equation of the diffusion parameter can be written as follows:

$$\alpha = 1.983 \times 10^{-3} \, e^{\left(\frac{-7439.7}{T}\right)} \tag{21}$$

4.3 Determination of the Rate-Controlling Step

The gasification process involves mass transfer and chemical reaction. To find out mechanisms which control this process, two approaches were developed, i.e., chemical reaction controls and diffusion through ash layer controls.

(a) Chemical reaction controls

If it is assumed that chemical reaction controls at the process of char gasification ($k_{TRZ} \lll \alpha$), Eq. (14) can be written in the form:

$$1 - (1 - X_B)^{\frac{1}{3}} = k_{TRZ} t \tag{22}$$

To obtained the value of k_{TRZ} in Eq. (22), linear regression analysis is conducted. The results are presented in Table 1.

The values of k_{TRZ} listed in Table 2 are used to calculate the carbon conversion and compared the result with experimental data. The deviation of this approach is presented in Fig. 9.

Figure 9 shows that by using chemical reaction control approach, the deviation of the calculation result toward the experimental data is relatively high. It can be seen from the fact that most of the intermittent symbols are not on a diagonal line.

Table 2 The value of k_{TRZ} at different temperatures

Temperature (°C)	The value of k_{TRZ} (min^{-1})	Lower (95% confidence)	Upper (95% confidence)
600	1.3011×10^{-4}	8.415×10^{-5}	1.76×10^{-4}
700	1.8718×10^{-4}	1.18×10^{-4}	2.57×10^{-4}
800	2.8212×10^{-4}	1.69×10^{-4}	3.95×10^{-4}

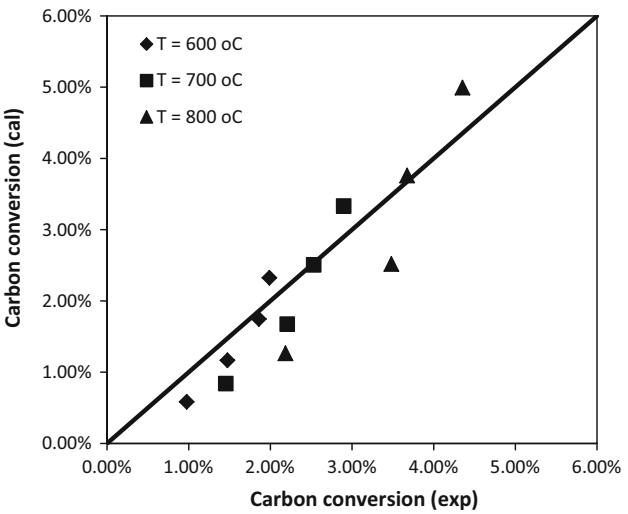


Fig. 9 The deviation of the calculated carbon conversion toward the result of experimental at temperature 600, 700, and 800 °C for 60 min using chemical reaction controls approach

The calculation resulted that the average of error at temperatures of 600, 700, and 800 °C were 16.79, 16.45, and 17.38% respectively. Thus, it can be concluded that chemical reaction is not controlling the process of gasification.

(b) Diffusion through ash layer controls

If it is assumed that diffusion through ash layer controls ($k_{TRZ} \ggg \alpha$), Eq. (14) can be written in the form:

$$\frac{1}{2}\left[1 - (1 - X_B)^{\frac{2}{3}}\right] - \frac{1}{3} X_B = \alpha t \tag{23}$$

Table 3 The value of α at different temperatures

Temperature (°C)	The value of α (min^{-1})	Lower (95% confidence)	Upper (95% confidence)
600	3.9208×10^{-7}	3.5319×10^{-7}	4.3097×10^{-7}
700	8.0771×10^{-7}	7.5149×10^{-7}	8.6393×10^{-7}
800	1.8269×10^{-6}	1.5728×10^{-6}	2.0809×10^{-6}

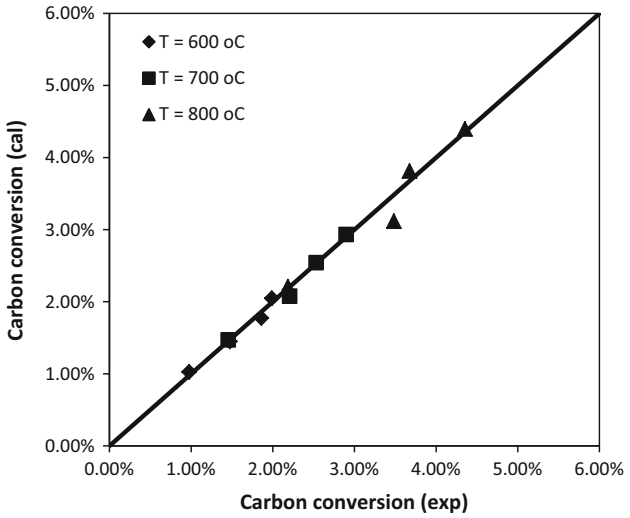


Fig. 10 The deviation of the calculated carbon conversion toward the result of experimental at temperature 600, 700, and 800 °C for 60 min using diffusion through ash layer controls approach

By using linear regression method, the value of α can be obtained. The results of the regression analysis are presented in Table 3.

Carbon conversion calculated using the value of α are listed in Table 3 and compared with the experimental data. The deviation of this approach is presented in Fig. 10.

From Fig. 10 it can be seen that by using diffusion through ash layer control approach, the deviation of the result of calculation is relatively small. It can be seen from the fact that most of the intermittent symbols are on a diagonal line.

The calculation showed that the average error at temperatures 600, 700, and 800 °C were 2.89, 1.75, and 3.26% respectively. Thus, the approach of the diffusion through ash layer control can describe the mechanisms of the process of gasification. As for the comparison between experimental data and the model, the result is presented in Fig. 11.

As shown in Fig. 11, the char conversion from the calculation of char conversion based on diffusion through ash layer controls approach fits well the experimental data. This result indicated that mechanisms of diffusion through ash layer controls in this process.

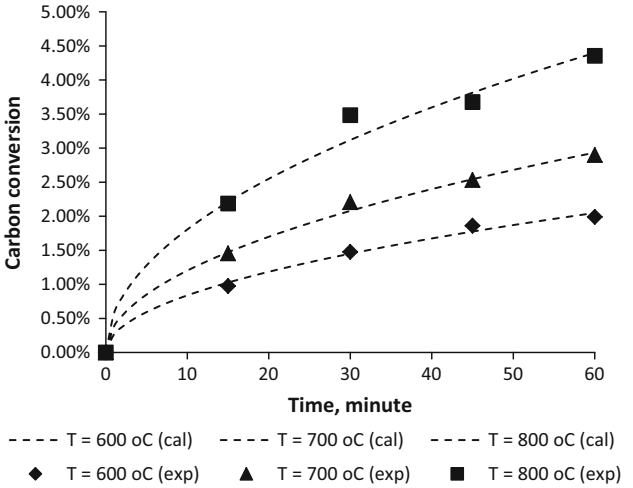


Fig. 11 Comparison of experimental data and calculated char conversion at steam gasification temperature 600, 700, and 800 °C for 60 min using diffusion through ash layer controls approach

5 Conclusions

Thin reaction zone model has ben presented and validated with experimental data. Data were obtained by conducting experiments on steam gasification of char at three different temperatures (600, 700, and 800 °C) for 60 min. Simulation has been done, and it can be concluded that the mechanisms of mass transfer through ash layer control the process of gasification.

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