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The Kinetics of CaO Assisted Pattukku Charcoal Steam Gasification

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Abstract

Coal is a solid fuel that can be converted into syngas through gasification process. To obtain optimum gasification process design and operation, in-depth understanding of the influential parameters is required. This study aims to investigate the effect of temperature on the gasification process and to obtain its kinetics parameters. The study was carried out in a tubular reactor equipped with a heater and a condenser. Steam was used as gasifying agent, while CaO was employed as a CO₂ adsorbent. The charcoal from coal was subjected to gasification at temperatures of 600, 700, and 800°C. The ratio of charcoal and CaO was 1:1. The gasification process lasted for 60 minutes with gas sample was taken every 15 minutes for composition analysis. The results showed that a temperature increase of 100°C caused a proportional increase of conversion of about 75% higher. The value of activation energy (*E_a*) and exponential factor (*k_o*) were 46.645 kJ/mole and 328.3894/min, respectively. For mass transfer parameters, values of activation energy for surface diffusion (*E_s*) and surface diffusivity factor (*α_s*) were 81.126 kJ/mole and 0.138/min, respectively.

Keywords: gasification; mathematical model; Pattukku coal char; steam; Thin Reaction Zone Model

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INTRODUCTION

Coal is one of alternative energy sources, which is still abundantly available in the world. One of the coal utilization technologies as a fuel source is by converting it into syngas through gasification process. In general, gasification is a thermal process for converting carbonaceous raw materials into flammable gases (Basu, 2010; Wang *et al.*, 2013; Heidenreich and Ugo, 2015; Demirbas, 2016).

Gasification is a complex process, so that good understanding on the influences of raw materials and other operating parameters is important for its design and operation process. To obtain such understandings,

a mathematical modeling can be used to perform the optimization of the process through the optimization of the influential parameters (Materazzi *et al.*, 2013; Babu and Sheth, 2006; Rezaiyan and Cheremisinoff, 2005). Although it could not provide very accurate results, the mathematical model often give some important clues about the occurring mechanisms and quantitative effects of design, raw materials, and other operating parameters (Baruah and Baruah, 2014; Basu, 2010; Molina and Mondrago, 1998; Xu *et al.*, 2011).

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The basic mechanism and the effect of kinetics parameters of a reaction are important information for the design of a gasification unit (Xu *et al.*, 2014;

Kopyscinski *et al.*, 2013; Baruah and Baruah, 2014; Zoulalian *et al.*, 2015). The use of reaction kinetics model to study the mechanisms and influential operating parameters in a gasification process has been done by several researchers. They developed some mechanistic models, such as Volumetric Model (VM), Shrinking Core Model (SCM), Random Pore Model (RPM) and Thin Reaction Zone Model (TRZM).

The VM uses the assumption that no structural changes occur during the gasification process. The gasifying agents are considered to be evenly distributed over the entire surface of the char particles. Then, it reacts with the active site of the char (Karimi *et al.*, 2011; Kraussler and Hofbauer, 2016). The reaction rate equation for this model is best expressed in the form of

$$-r_A = \frac{dX_A}{dt} = k(1 - X_A) \quad (1)$$

The SCM uses the assumption that the reaction takes place on the pore surface of the solid particles or on the surface of a non-porous solid. The gasification reaction rate according to SCM mechanism can be represented by (Zhang *et al.*, 2010; Adanez and Diego, 1990; Levenspiel, 1999)

$$-r_A = \frac{dX_A}{dt} = k(1 - X_A)^{\frac{2}{3}} \quad (2)$$

The RPM was proposed by Bhatia and Perlmutter. This model assumes that the char consists of cylindrical pores with random size distributions (Bhatia and Perlmutter, 1980; Zhang *et al.*, 2010; Tang *et al.*, 2015). The reaction rate is then expressed in the form of

$$-r_A = \frac{dX_A}{dt} = k_p(1 - X_A)\sqrt{1 - \Psi \ln(1 - X_A)} \quad (3)$$

The RPM is more flexible than both VM and SCM. It is mostly used in the modeling of coal or carbon charcoal gasification (Kajitani *et al.*, 2006; Everson *et al.*, 2008). However, the RPM produces less satisfactory results for charcoal which has rather complex structures, such as biomass or low rank coal (Goyal *et al.*, 1989; Hurt *et al.*, 1986).

The TRZM was developed based on the assumption that the mass transfer rate of the gas reactants in the film layer around the solid granules is not the rate controlling mechanism. This assumption is supported by the fact that the reactions occur on the very thin grain surfaces (Syarif *et al.*, 2017). This model can well describe the process of gasification of low rank coal. The TRZM model can be written as the following

$$t = \left(\frac{1}{k_{TRZ}} \right) \left[1 - (1 - X_B)^{\frac{1}{3}} \right] + \left(\frac{1}{\alpha} \right) \left[\frac{1}{2} - \frac{1}{2}(1 - X_B)^{\frac{2}{3}} - \frac{1}{3}X_B \right] \quad (4)$$

This study aims to study the influence of temperature in the gasification process and to obtain the value of the reaction kinetics constants (chemical reaction and mass transfer). The value of each parameter is obtained through simulation process of observation data using TRZM model.

MATERIALS AND METHODS

Materials

The raw materials used in this study were coal, steam, Nitrogen gas (N₂), and limestone. Pattukku Coal, which was obtained from Bone regency of South Sulawesi, was used as raw material for the preparation of charcoal. This coal has high sulfur content, so that it falls in the low rank coal category. The results of coal proximate analysis are presented in Table 1.

Table 1. Proximate analysis results of Pattukku coal

Component	Composition (% weight)
Volatile compounds	38.28
Carbon remains	47.92
Humidity	4.70
Ash	10.80

Research Apparatus

The main apparatus used in this research were furnace and tubular reactor made from quartz with 3.5 cm diameter and 45 cm height dimension. The auxiliaries, such as condenser, condensate tube, syringe, and gas product container tube were also used in this study. The equipment was presented in Figure 1.

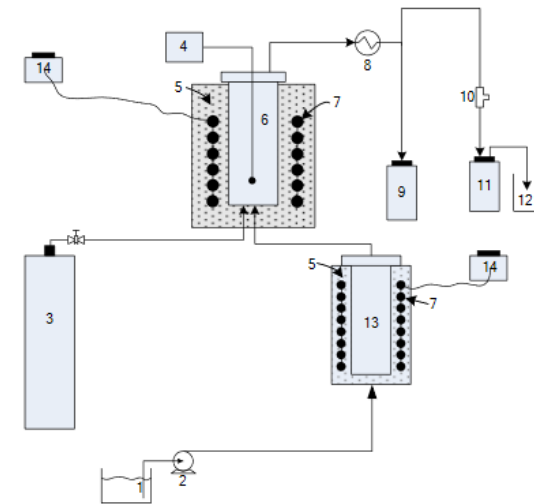


Figure 1. The experimental set-up
(1. Water tank, 2. Pump, 3. N₂ tank, 4. Thermocouple, 5. Isolator, 6. Reactor, 7. Electric heater, 8. Condenser, 9. Condensate tank, 10. Sampling gas, 11. Water tank/gas container, 12. Water container, 13. Steam maker, 14. Regulator)

Charcoal Preparation

The charcoal was prepared by pyrolysis of Pattukku coal to remove the existing volatile compounds. Two hundred grams of coal having 3.35-4 mm particles diameter was introduced into the reactor. The N₂ gas was passed through the reactor from its bottom entrance for 15 minutes to remove the air initially contained in the reactor. Furnace was turned on

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$$t = \left(\frac{1}{k_{TRZ}} \right) \left[1 - (1 - X_B)^{\frac{1}{a}} \right] + \left(\frac{1}{a} \right) \left[\frac{1}{2} - \frac{1}{2} (1 - X_B)^{\frac{2}{a}} - \frac{1}{3} X_B \right] \quad (4)$$

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to heat up the reactor to reach temperature of 450°C. When the desired temperature was reached, the temperature control device was turned on so that the pyrolysis took place isothermally. After 60 minutes, the electric heater was turned off and the reactor temperature was allowed to equilibrate to room temperature (30°C). The composition of the formed charcoal was analyzed using ultimate analysis at TEKMIIRA Laboratory Bandung. The results of the ultimate analysis are presented in Table 2.

Table 2. The ultimate analysis results

Component	Composition (% weight)
C	68.93
H	2.99
O	24.39
N	1.81
S	1.88

Calcination of Limestone

The limestone, which was used in this study, was calcined first to break down the existing CaCO_3 into CaO. A total of 200 grams of limestone samples were put into the muffle furnace. Furnace was operated at 900°C with holding time of 3 hours. After 3 hours, the furnace was turned off and was left to equilibrate with ambient air temperature of 30°C. The calcined limestone was then analyzed using LDX-8000 (Energy Dispersive X-ray Fluorescence Spectrometer) at the FT-Chemical Engineering Data and Instrumentation Analysis Laboratory. The results of the analysis are presented in Table 3.

Table 3. The ultimate analysis results

Component	Composition (% weight)
CaO	94.012
SiO_2	2.814
Al_2O_3	1.59
SO_3	1.013
Impurities	0.571

Charcoal Gasification

A mixture of coal and calcined lime with a ratio of 1:1 was fed into the reactor. The N_2 gas was flown from the bottom part of the reactor for 15 minutes to remove air from the reactor. The electric heater (furnace) was turned on to raise the temperature of the reactor until it reached the desired reaction temperature. When the reaction temperature was reached, the steam was passed through the reactor to via its bottom entrance for reaction with the charcoal. The gasification process took place at a fixed temperature for 60 minutes and every 15 minutes sampling was taken using syringes for gas analysis. The composition of the gases (H_2 , CH_4 , CO , and CO_2) were analyzed using a gas chromatography (GC) Shimadzu GC-2010. The experiment was repeated at three different temperatures, i.e. 600, 700, and 800°C. The reaction conversion can be calculated based on the gas composition and gas volume at various time by the following equation

$$X_C = \frac{n_{\text{CH}_4} + n_{\text{CO}} + n_{\text{CO}_2}}{n_{\text{CO}}} \times 100\% \quad (5)$$

Mole of C in the initial sample (n_{CO}) was calculated using equation (6)

$$n_{\text{CO}} = \frac{0.6893 \times m_{\text{char}}}{12} \quad (6)$$

RESULTS AND DISCUSSION

Effect of Temperature

Figure 1 shows that the conversion of the gasification process increases with time. At temperatures of 600°C and 700°C, the conversion increases linearly with time up to 45 minutes, then it increases gradually until the end of the experiments. Different phenomenon was observed at higher temperature where the conversion steadily increases with time. The conversion of the reaction was also found to increase in line with the increasing temperatures. At 600°C, the final conversion obtained was 4%. It is clearly observed that the conversion increases about $\frac{3}{4}$ times when the temperature was increased by 100°C. This conversion increase with respect to the increase in temperature was likely to be due to the increment of the kinetics energy of the molecules, which led to the highly intensive collisions between reacting particles. A higher temperature will promote a greater reactivity of charcoal and triggers faster reaction rate and the formation of larger amount of syngas (Hu *et al.*, 2006; Mahishi and Goswami, 2007). Some previous researchers also showed the same trend. The comparison between the results of this study with other previous researches is presented in Table 4.

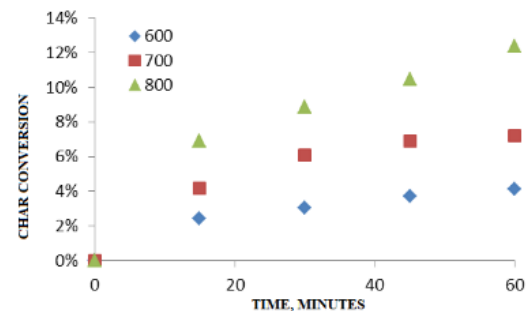


Figure 1. The relationship of charcoal vs time conversion at various reaction temperatures

Table 4. Comparison of results with previous researchers

Researchers	Raw material	Conversion increase	Average hicks
Murakami <i>et al.</i> , (2015)	Coal	1.9 (700 to 750°C)	0.035/°C
Mahishi and Goswami (2007)	Adaro	1.7 (750 to 800°C)	
	Pine bark	1.3 (500 to 600°C)	0.013/°C
		1.3 (600 to 700°C)	
This research	Pattukku charcoal	1.8 (600 to 700°C)	0.018/°C
		1.7 (700 to 800°C)	

reactor until it reached the desired reaction temperature. When the reaction temperature was reached, the steam was passed through the reactor to via its bottom entrance for reaction with the charcoal. The gasification process took place at a fixed temperature for 60 minutes and every 15 minutes sampling was taken using *syringes* for gas analysis. The composition of the gases (H_2 , CH_4 , CO , and CO_2) were analyzed using a gas chromatography (GC) Shimadzu GC-2010. The experiment was repeated at three different temperatures, i.e. 600, 700, and 800°C. The reaction conversion can be calculated based on the gas composition and gas volume at various time by the following equation

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		1.7 (700 to 800°C)	

Kinetics Model Validation

The kinetics model was validated using the curve fitting method by minimizing the value of Sum Square of Error (SSE). From the minimization of SSE during the simulation process the optimized kinetics parameter values (k_{TRZM}) and mass transfer (α) can be obtained. The comparison of the conversion at various reaction time obtained from the experiment and calculation results is presented in Figure 2.

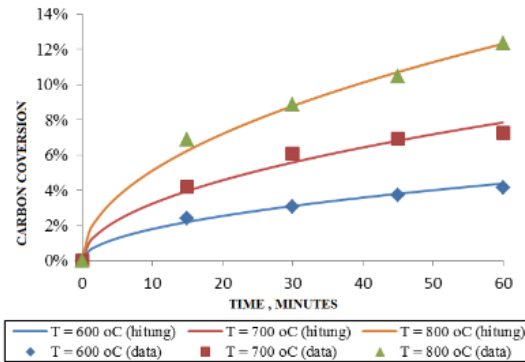


Figure 2. Comparison of observed conversion results with calculations at various temperatures

Figure 2 shows that the calculated charcoal conversions agree well with the experimental data. The obtained average errors for temperatures of 600, 700, and 800°C are 2.86; 4.15; and 3.19%, respectively. Visually distorted calculations results with observational data are presented in Figure 3.

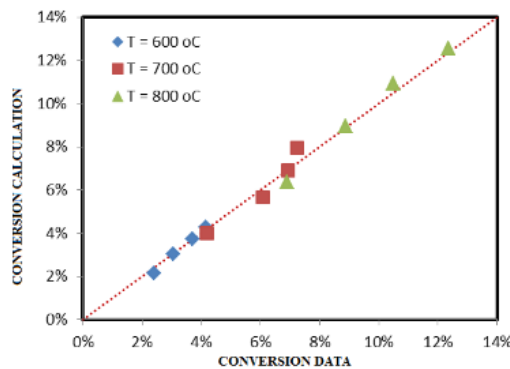


Figure 3. Deviation of conversion of calculation results with observation data at various temperatures

Figure 3 shows that almost all of the points that reflect the conversions obtained from experiment and calculation fall on the diagonal line. It shows that the deviation between data conversion and calculation is very small. From the obtained calculation results, the correlation coefficients (R^2) for temperatures of 600, 700, and 800°C each are 0.998; 0.971; 0.996. Thus, it can be said that TRZM can be used to explain the occurring mechanisms with high accuracy.

Kinetics Constants Evaluation (k_{TRZM})

Based on the k_{TRZM} value obtained from the simulation process, a plot of the relationship between $\ln k_{TRZM}$ versus $1/T$ is shown in Figure 4.

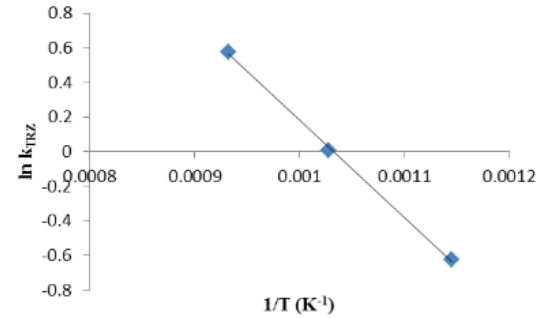


Figure 4. Relationship between $\ln k_{TRZM}$ versus $1/T$

By plotting a straight line through the three points, the *slope* ($-E_a/R$) and *intercept* ($\ln k_{TRZM}$) can be obtained. Their values are -5610.4 and 5.7942, respectively. Thus, the correlation between k_{TRZM} and temperature can be written as follows

$$k_{TRZ} = 328.3894 \exp\left(-\frac{46644.87}{RT}\right) \quad (7)$$

Mass Transfer Period Evaluation (α)

The mass transfer constant (α) as a function of temperature can be approximated by the following form (Perry, 2008)

$$\alpha = \alpha_s \exp\left(-\frac{E_s}{RT}\right) \quad (8)$$

The value of α_s and E_s can be obtained by plotting $\ln \alpha$ versus $1/T$. A straight-line should connect all the data. Then, the *slope* ($-E_s/R$) and *intercept* ($\ln \alpha_s$) can be obtained. The values of α_s and E_s were respectively 0.138 minutes⁻¹ and 81125.5 J/mole. Accordingly, the α as a function of temperature can be written as

$$\alpha = 0.138 \exp\left(-\frac{81125.5}{RT}\right) \quad (9)$$

CONCLUSION

Gasification is a complex thermal process because it involves simultaneous physical and chemical processes. Temperature is one of the important factors in the process of gasification. By the increasing temperature into 100°C, the reaction conversion increases $\frac{3}{4}$ times from the previous conversion. From the simulation process that has been done using Thin Reaction Zone Model, the activation energy value (E_a) and exponential factor (k_o) respectively are 46.645 kJ/mole and 328.3894min⁻¹. For the parameter of mass transfer, the values that are obtained from activation energy for surface diffusion (E_s) and surface diffusivity factor (α_s) are 81.126 kJ/mole and 0.138 min⁻¹.

0% 2% 4% 6% 8% 10% 12% 14%
CONVERSION DATA

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