Modeling Product Distribution of Top-lit Updraft Gasification

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A kinetic model for predicting biochar, producer gas, and tar formations of top-lit updraft (TLUD) gasification was developed. The three main zones within the TLUD gasifier, the pyrolysis, incomplete combustion, and reduction reaction zones, were incorporated into the model and sequentially solved. Validated with experimental data, the model was found capable of predicting biochar yield on pine woodchips at varying airflow rates, biomass moisture contents, and biomass compactness. However, when the particle size was varied, the model underestimated biochar yield. The model also accurately predicted the higher heating value of the producer gas that varied from 3.45 to 3.98 MJ/m³ compared to 3.61 to 3.67 MJ/m³ for the experimental results. The model qualitatively predicted tar content in the producer gas at varying conditions. However, accurate quantification of tar generation in TLUD gasification was not achieved.

Keywords: Top-lit updraft; Gasification; Kinetic model; Biomass; Producer gas

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INTRODUCTION

Agricultural residues and municipal wastes are some examples of raw materials that are used worldwide to produce energy and biomaterials (Yahya *et al.* 2015) through various processes such as chemical, biological, and thermal conversions (Azman *et al.* 2015; Kumar *et al.* 2015; Raheem *et al.* 2015). As a popular thermal conversion technology, biomass gasification is an incomplete oxidation of biomass that produces syngas (or producer gas) and biochar (Gómez-Barea and Leckner 2010). Syngas is a mixture of H₂, CO, CH₄, and some non-combustible gases that can be used to generate electricity, or converted into hydrocarbons *via* the Fischer Tropsch process (Jameel *et al.* 2010). Biochar is a carbon-rich material that can improve water and nutrient retention in soils, absorb pollutants, and produce H₂ *via* steam reforming reactions (Winsley 2007; James *et al.* 2014). Although gasifiers can produce biochar, they are designed and optimized for making gas products that result in nearly complete utilization of the carbon in biomass (Sharma 2008). There is not extensive work focused on the production of biochar by gasification processes (Brown 2009).

Top-lit updraft (TLUD) biomass gasifiers produce relatively high yields of biochar when compared to other gasification units such as fluidized bed, downdraft, and updraft gasifiers (Nsamba *et al.* 2014). A top-lit updraft gasifier is a batch gasifier that is known to be ignited on the top layer of the biomass. Because of the flow of the biomass and the gasifying agent, it is also known as an inverted downdraft gasifier. The use of volatiles to

drive the thermochemical reactions make this gasifier useful to generate heat with relatively high efficiency. For this reason, the TLUD configuration is often used to build stoves in developing countries. The TLUD configuration has been reported to discourage the formation of tar at high temperatures (825°C). Therefore, the reduction in tar content can be associated with the combustion of tar in the combustion zone. The tar reduction can also be attributed to the catalytic effect of the top layer of carbon that remains at a relatively high temperature. In addition, the top layer of biochar could also be responsible for a slight increase in hydrogen composition and HHV (higher heating value) of the syngas because of a more stable reduction zone in a carbon-rich environment (James et al. 2014). However, current literature has paid little attention to simultaneous production of syngas and biochar from this type of reactor. Huangfu et al. (2014) studied the effect of moisture content on the thermal performance and emissions of a TLUD gasifier, but the potential use of the produced biochar was not addressed. Similarly, Saravanakumar et al. (2007) studied the production of syngas from long stick wood in a TLUD gasifier. However, the production of biochar from this process was not considered. Recent work of the authors has presented the potential of TLUD gasification for biochar and producer gas generation (James et al. 2018). The highest biochar yield achieved was 39%, and the highest gas concentrations were 6.6% (v/v) and 15% (v/v) for H_2 and CO in the producer gas, respectively.

Extensive work has been reported on the prediction of syngas production from biomass gasification (Yang *et al.* 2004; Gøbel *et al.* 2007; Puig-Arnavat *et al.* 2010). Several approaches have been implemented. These include equilibrium, kinetic, and computational models (*e.g.*, ASPEN, Computational fluid dynamics, *etc.*) (Gómez-Barea and Leckner 2010; Patra and Sheth 2015). Syngas composition, tar content, and carbon conversion are often considered in the modeling process (Tinaut *et al.* 2008). However, these models do not consider biochar as a product of gasification. They are rather associated with the prediction of conditions at which low gasification efficiency is expected (Fiaschi and Michelini 2001). In contrast, pyrolysis models can effectively predict the yield of biochar from a variety of biomasses (Koufopanos *et al.* 1989; Sharma *et al.* 2015). However, they are unsuitable for the prediction of biochar production from biomass gasification because of the oxygen-free nature of the pyrolysis process that is contrary to the thermochemical conversion of biomass gasification. As a result, a model that considers both biochar and producer gas as products of gasification is needed.

The goal of this work is to develop a kinetic model for the prediction of the products of TLUD biomass gasification. The formation of biochar, producer gas, and tar were considered in the model at varying airflow rates, biomass particle sizes, moisture contents, and biomass compactness. This approach could help identify the expected output of the gasifier at different conditions.

EXPERIMENTAL

Modeling Considerations

The present model considers the chemical as well as physical properties of the biomass. The product distributions of pine woodchips gasification were evaluated at 8, 12, 16, and 20 L/min airflow rates, corresponding to 0.016, 0.024, 0.032, and 0.040 m/s superficial velocities, respectively. Table 1 presents the proximate and ultimate analyses of the pine woodchips that were tested. The experimental setup for validation and gasification methodology for varying airflow rates was presented elsewhere (James *et al.*)

2018). The evaluation of the physical properties for woodchips included four levels of moisture content (10, 14, 18, and 22%), particle size (2, 7, 17, and 30 mm), and biomass compactness (0, 1, 2, and 3 kg). The experimental procedures and experimental results were previously reported (James *et al.* 2016).

C (%)	H (%)	N (%)	O (%)	S (%)	Ash (%)	Volatile Matter (%)	Fixed Carbon (%)	Moisture Content (%)	Bulk Density (g/cm ³)
47.90	1.70	0.30	49.90	0.20	0.57	74.92	16.66	7.85	2.11

Table 1. Proximate and Ultimate Analyses of Pine Woodchi	ips
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The following assumptions were considered when modeling the TLUD biomass gasifier:

- 1. The tar fuels the incomplete combustion reactions, and they occur instantaneously.
- 2. The concentrations of reactants and products do not vary in the radial direction of the tubular reactor. The reactor operates in isothermal and adiabatic steady state mode.
- 3. The pressure differential in the reactor is negligible.
- 4. Ash does not react during thermochemical reactions because the reaction temperature is below 1,000 °C.
- 5. All gases are considered ideal gases. Biochar is considered pure carbon.
- 6. Heterogeneous reactions are not reversible.

The present model considers three main stages of biomass reaction, an instantaneous devolatilization of biomass, the incomplete combustion of tar, and the reduction reactions. In the first two stages, the reactions were considered instantaneous. In addition, the heterogeneous solid/gas reactions were evaluated with a kinetic model. The details are described as follows.

In a TLUD gasifier, the flaming pyrolysis is displaced from the top to the bottom of the gasifier as the reactions take place. This gives the gasifier a characteristic configuration in which the combustion zone provides the needed heat for pyrolysis and reduction reactions to take place (Fig. 1).



Fig. 1. Gasification zone distribution of a top-lit updraft gasifier. As the biomass reacts, the incomplete combustion zone moves towards the bottom of the gasifier, while the gasification agent (air) flows upwards in the fixed-bed batch reactor.

The heat from the combustion zone devolatilizes the biomass as part of the pyrolysis reactions (Saravanakumar *et al.* 2007). Moreover, on the top of the combustion zone, biochar and gases are generated due to reduction reactions (Patra and Sheth 2015). Therefore, the present model considered these three major zones in the following order: pyrolysis, incomplete combustion, and reduction zones. The pyrolysis reactions were represented by the molar balance of Eq. 1 in which carbon (biochar), CO₂, CH₄, and tar (CH_{1.03}O_{0.03}) are the products of the thermal decomposition of biomass (pyrolysis) at high temperatures (Font Palma 2013),

$$CH_{\alpha}O_{\beta} \xrightarrow{\Delta T} x_1C + x_2CO_2 + x_3CH_4 + x_4CH_{1.03}O_{0.03} \tag{1}$$

where α and β are the moles of hydrogen and oxygen per mole of carbon in the biomass, respectively. The stoichiometric coefficients x_1 , x_2 , x_3 , and x_4 are the moles of individual products generated during the pyrolysis of biomass.

The initial yield of biochar was calculated based on the maximum amount of biochar generated when considering the carbon yield from cellulose, hemicellulose, and lignin as presented by Sharma (2011). Moreover, the incomplete combustion of tars takes place with the corresponding air/fuel ratio for the studied airflow rates. Equation 2 shows the reaction of the incomplete combustion of tar that results in CO and H₂ (Tinaut *et al.* 2008):

$$CH_{1.03}O_{0.03} + 0.483 O_2 \rightarrow CO + 0.517 H_2$$
(2)

The products from the first two stages were used to feed the reduction reactions. Heterogeneous (solid/gas) reactions of the biochar and the gases from previous reactions were considered and are presented in Eqs. 3 through 5. Similar approaches for the reduction zone have been widely implemented in the literature (Giltrap *et al.* 2003; Babu and Sheth 2006; Zhong *et al.* 2009). Equations 3 through 5 are as follows:

 $C + CO_2 \rightarrow 2CO$ (3)

$$C + 2H_2 \rightarrow CH_4 \tag{4}$$

$$C + H_2 O \rightarrow CO + H_2 \tag{5}$$

Despite the fact that TLUD gasification is often associated with downdraft biomass gasification (Pérez *et al.* 2012), the characteristic reaction mechanisms of this reactor enable it to produce biochar. Reed *et al.* (1988) presented an empirical equation for the prediction of the flaming pyrolysis time experienced by a biomass particle in a TLUD gasification unit. Equation 6 accounts for the moisture content, biomass density, reaction temperature, molar fraction of oxygen in the air, and shape of the biomass particles. In this model, because the time for incomplete combustion of tars is considered instantaneous, the time for flaming pyrolysis time is calculated according to Eq. 6,

$$t_{\rm fp} = (0.207 \ \rho \ F_{\rm s} \ D \ (1+1.76 \ F_{\rm m}) \ (1+0.61D) \ e^{(9369/RT)})/(1+3.46 \ Fo_2) \tag{6}$$

where $t_{\rm fp}$ is the flaming pyrolysis time (min), ρ is the density of the biomass particle (g/cm³), $F_{\rm s}$ is the sphericity of biomass materials (for biomass materials, $F_{\rm s} = 0.2$, (de Souza-Santos 2010), D is the diameter of biomass particle (cm), $F_{\rm m}$ is the weight fraction of moisture content in biomass (g/g), R is the ideal gas constant (J/mol/K), T is the temperature (K), and Fo_2 is the molar fraction of oxygen in the gasification agent (0.21 for air).

The reaction rates for the kinetic model are defined in Eqs. 7 through 10. The reaction rate for the incomplete combustion of tar (Eq. 7) was adopted from Tinaut *et al.* (2008). Moreover, the reaction rates for the reduction reactions were calculated based on reactions that obey the elementary rate law (Fogler 2006). These equations were considered irreversible (Eq. 8 and 9),

$$r_{\rm Com} = Tk_{\rm t}C_{\rm char}C_{\rm t}^{0.5} \tag{7}$$

$$r_{\rm gas1} = k_1 C_{\rm char} C co_2 \tag{8}$$

$$r_{\rm gas2} = k_2 C_{\rm char} C_{\rm H2O} \tag{9}$$

$$r_{\text{gas3}} = k_3 C_{\text{char}} C_{\text{H2}} \tag{10}$$

where r_{Com} is the reaction rate for the incomplete combustion of biomass (Eq. 2); r_{gas1} , r_{gas2} , and r_{gas3} are the reaction rate for Eqs. 3, 4, and 5, respectively. *T* is the reaction temperature in Kelvin. k_t , k_1 , k_2 , and k_3 are the kinetic constants for the Eqs. 2, 3, 4, and 5, respectively. *C*_{char}, *C*_t, *C*_{CO2}, *C*_{H2O}, and *C*_{H2} are the concentrations of carbon, tar, carbon dioxide, water, and hydrogen in moles/m³, respectively.

The kinetic constants k_i were calculated according to Arrhenius equation (Eq. 11) that considers the temperature dependence of chemical reactions (Fogler 2006),

$$k_{\rm i} = A_{\rm i} e^{-{\rm Ei}/{\rm RT}} \tag{11}$$

where k_i is the kinetic constant for Eqs. 2, 3, 4, and 5, *A* is the pre-exponential factor (1/s), E_i is the activation energy of the specific reactions (kJ/mol/K), *T* is the reaction temperature (K), and *R* is the ideal gas constant (KJ/mol). Table 2 presents the activation energy and pre-exponential factors for the reaction rates.

Reaction A (1/s)		E (KJ/mol/K)	Reference		
Tar	59.8	101.43	(Déroz 2007: Tipout et al. 2008)		
gas1	3.42 T	129.7	(Perez 2007, Tillaut <i>et al</i> . 2008)		
gas2	4.18E-3	129.7			
gas3	7.301E-2	129.7	(Wang and Kinoshita 1993; Perez 2007; Tinaut <i>et al.</i> 2008)		

The differential equation below describes the production and disappearance of carbon, gases, and tar due to incomplete combustion and reduction reactions. They were defined based on the fundamental Eq. 12 for batch reactors (Speight and Özüm 2002),

$$dC_{j}/dt = \sum_{i=1}^{R} \alpha_{i,j} r_{i}$$
(12)

where C_j (moles/m³) is the concentration of a substance (j = Carbon, H₂, CO, CH₄, CO₂, H₂O, or tar), $\alpha_{i,j}$ is the stoichiometric coefficient for corresponding reactant/product, r_i is the reaction rate for individual reactions, and t (s) is the reaction time.

The differential equations for tar combustion and reduction reactions were solved in MATLAB® (MathWorks, version R2015a, Natick, MA, USA) with the integrated ordinary differential equation solver ODE15s. The biomass carbon concentration within the gasifier and the concentration of oxygen in the gasification agent were used as the initial reactants in the tubular reactor. Then, the outputs of modeling the incomplete combustion of tars were used as the initial reactants for the reduction reactions.

RESULTS AND DISCUSSION

Model Validation

The predicted concentrations of the products were validated with the experimental results presented in previous work (James *et al.* 2016, 2018). Figure 2 shows the yield of biochar at varying airflow rates, which decreased as the airflow rate increased. This trend was also presented by the model in close approximation to the experimental data. The corresponding air-fuel (A/F) ratios for the gasification airflows of 8, 12, 16, and 20 L/min were 1.22, 1.44, 1.55, and 1.73, respectively. These could be compared with the A/F for theoretical complete combustion of the biomass of 10.37. The relatively low A/F ratios for the gasification in addition, the downward movent of the combustion layer resulted in the generation of biochar.





Table 3 presents the predicted and experimental results of higher heating value (HHV) and tar content in the producer gas. Model prediction of HHV was in close agreement with experimental data, with maximum prediction error of 8.8% (at 8 L/min) and average absolute prediction error of 4.5%. The predicted tar contents show a consistent trend with the experimental data; however, actual values were way off especially at low airflow, which suggests that the model is less accurate at lower airflow for tar prediction.

	Experiment (James <i>et</i>	al Results <i>al</i> . 2018)	Modeling Results	
Airflow Rate (L/min)	HHV (MJ/m ³)	Tar (g/m ³)	HHV (MJ/m ³)	Tar (g/m³)
8	3.17	86.24	3.45	26.13
12	3.50	49.30	3.42	20.88
16	3.57	22.19	3.77	17.85
20	3.93	12.99	3.98	15.25

Table 3. Experimental and Modeling Results of Syngas HHV and Tar Content at

 Varying Airflow Rates*

*Particle size 2 mm, moisture content 10%, and biomass compactness 0 kg

Effects of Particle Size

The comparison of the predicted and experimental yield of biochar at different particle sizes is shown in Fig. 3. The predicted biochar yield presented a similar trend as that exhibited by the experiments. However, in most cases the model under-predicted biochar yield. This could be related to the fact that the reactor was assumed to be isothermal. As a result, an overall higher temperature was presented in the reduction zone that led to the utilization of more carbon in the solid/gas reactions (Wang and Kinoshita 1993; Babu and Sheth 2006). In addition, the reduction in biochar yield for particle size change from 3 to 7 mm and the increase in yield for particle sizes larger than 7 mm were previously attributed to the reaction temperature that changed due to the change in biomass density. The effect of the physical properties of biomass on top-lit updraft gasification was presented by the authors elsewhere (James *et al.* 2016).





The prediction of the HHV and tar content when varying the particle size are shown in Table 4. Despite the adjustment of the flaming pyrolysis time at different particle sizes in the model, the tar contents were still underestimated in most cases, although the predicted tar content *vs.* particle size followed a similar decreasing/increasing trend as the experimental values, indicating that the model was qualitatively accurate in predicting tar yield with varying biomass particle sizes. The model performed a better estimation of the HHV of the producer gas with an average absolute prediction error of 8.8%.

	Experime	ntal Results	Modeling Results			
	(James e	et al. 2016)	Wodoling Roodito			
Particle Size (mm)	HHV (MJ/m ³)	Tar (g/m³)	HHV (MJ/m ³)	Tar (g/m ³)		
2	3.26	79.43	3.22	17.13		
7	3.67	12.99	3.98	15.25		
17	2.94	69.71	3.49	19.52		
30	2.71	93.51	2.90	21.42		

Table 4. Experimental and Modeling Results of Syngas HHV and Tar Content at Varying Particle Sizes*

*Airflow rate 20 L/min, moisture content 10%, and compactness 0 kg

Effects of Moisture Content

The addition of moisture to biomass reduced biochar yield (Fig. 4). This tendency was also described by the model. However, at moisture contents higher that 18%, the biochar yield was underestimated by the model. This discrepancy could be related to the fact that the increase in biomass moisture content increased the flaming pyrolysis time because additional energy was needed to devolatilize the biomass particles. As a result, the produced carbon was longer exposed to reduction reactions. Thus, the yield of biochar further decreased (Huangfu *et al.* 2014).



Fig. 4. Biochar yield of woodchips at different levels of moisture content (airflow rate 20 L/min, particle size 7 mm, and compactness 0 kg)

The model over-predicted the HHV of the producer gas generated when moisture was added to the biomass (Table 5). Despite this, it qualitatively predicted the yield of tar with respect to moisture content, with the largest difference of 2.84 g/m^3 when the moisture content was 14%. This difference could be a result of the additional carbon used in the reduction reactions due to the increase in the flaming pyrolysis time.

	Experimer	nt Results	Modeling Results	
	(James et	<i>al</i> . 2016)		
Moisture Content (%)	HHV (MJ/m ³)	Tar (g/m ³)	HHV (MJ/m ³)	Tar (g/m ³)
10	3.67	12.99	3.98	15.25
14	3.13	7.56	3.76	10.40
18	2.65	6.18	3.84	8.39
22	2.84	6.24	3.85	6.37

Table 5. Experimental and Modeling Results of Syngas HHV and Tar Content at Varying Moisture Contents*

*Airflow rate 20 L/min, particle size 7 mm, and compactness 0 kg

Effects of Biomass Compactness

The biochar yield at different biomass compactness levels is presented in Fig. 5. The increase in biochar yield when the biomass compactness increased was accurately predicted by the model. However, the model under-predicted the yield of biochar by 1.86% points at biomass compactness of 3 kg. This could be related to the increase in the bulk density of biomass within the chamber because the reaction temperature did not have a noticeable change when the biomass compactness increased from 2 to 3 kg.



Fig. 5. Biochar yield of woodchips at different levels of compactness (airflow rate 20 L/min, particle size 7 mm, and moisture content 10%)

Table 6 shows the experimental and predicted HHV and tar content with varying biomass compactness. The model was able to accurately predict the HHV of the producer gas with a difference of 0.31 MJ/m^3 with the experimental data. However, it underpredicted the tar content at high levels of compactness. For instance, the tar content presented a difference of 30.64 g/m^3 between experimental and predicted result with 3 kg of biomass compactness. This under-prediction of the tar could be attributed to the assumption that only tar fueled the combustion reaction, instead of part of the produced carbon resulting of the pyrolysis process. As a result, the presented model depletes the tar in the combustion.

	Experiment (James <i>et</i>	al Results <i>al</i> . 2016)	Modeling Results	
Packing Mass	HHV	Tar	HHV	Tar
(kg)	(MJ/m³)	(g/m ³)	(MJ/m ³)	(g/m ³)
0	3.67	12.99	3.98	15.25
1	3.49	24.87	3.37	14.61
2	3.52	20.42	3.31	15.00
3	3.61	47.51	3.45	16.87

Table 6. Experimental and Modeling Results of Syngas HHV and Tar Content at

 Varying Biomass Compactness*

*Airflow rate 20 L/min, particle size 7 mm, and moisture content 10%

Overall Biochar Yield

Figure 6 shows the predicted and experimental values of biochar yield when the airflow rate, particle size, moisture content, and biomass compactness were considered. It was evident that an accurate prediction of biochar yield was achieved by the model in the lower to the mid-range. However, at high experimental yields, the model under-predicted their values. James *et al.* (2018) found a strong correlation of the airflow rate with the peak reaction temperature ($R^2 = 0.99$) and biochar yield ($R^2 = 0.93$). The variation of the temperature at different conditions plays a crucial role on biochar yield, which decreases

as the reaction temperature increases (Demirbas 2001, 2004; Sun *et al.* 2014; James *et al.* 2016). Therefore, it can be stated that the present model can effectively predict the yield of biochar at high reaction temperatures (low biochar yield). However, it lacks precision at low reaction temperatures, which can be caused by a small airflow, high moisture content, large biomass compactness, or extreme particle size (too small or too big).



Fig. 6. Comparison of predicted and measured yield of biochar

CONCLUSIONS

- 1. A model for predicting the products of top-lit updraft batch reactor for biomass gasification was developed and validated with experimental data. The model considered an adiabatic-isothermal reactor that experienced three stages of biomass conversion: fast devolatilization, tar combustion, and reduction reactions. MATLAB's ODE15s differential equation solver was implemented to solve the tar combustion and reduction reactions equations.
- 2. The model showed accurate prediction of biochar yield of pine woodchips at different airflow rates, moisture contents, and compactness of biomass. However, when the particle size varied, the model underestimated the yield of biochar.
- 3. The model could also predict the higher heating value of the producer gas at different airflow rates, particle sizes, moisture contents, and biomass compactness.
- 4. The model qualitatively predicted tar content in the producer gas at varying conditions. However, accurate quantification of tar yield was not achieved.

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