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# Numerical Study of Torrefaction-Gasification Integrated Process

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#### Abstract

Torrefaction as pretreatment for the thermochemical conversions is one of the profound technologies. In this work, torrefaction-gasification integrated process has been modeled and studied. First, a combination of different torrefaction models was adopted to quantify the ultimate analysis of the torrefied biomass. Then, the torrefied biomass was set as the input to gasification model and the results of gasification were analyzed. Impacts of different torrefaction conditions on gasification were evaluated. With torrefaction-gasification integrated process, composition of syngas was improved. The increase in severity of torrefaction conditions, enhanced the quality of syngas.

**Keywords:** Torrefaction; Gasification; Integrated model; Thermochemical conversion

# Introduction

Renewable energy is one of the solutions to meet the increasing requirements of energy with the depletion of fossil fuels' quantity [1]. Biomass is the potential source of renewable energy. However, biomass has some issues when used as fuel. Some of the issues related with biomass include low energy density, high moisture content and high O/C ratios.

For the thermochemical conversions of biomass, some pretreatment techniques have been developed including: drying, fast pyrolysis, slow pyrolysis, pelletization and torrefaction. Torrefaction is one of the promising pretreatment techniques used for thermochemical processes [2]. Researchers presented a comprehensive review on torrefaction and its applications [1].

Researcher compared the impacts of different four pretreatment techniques (drying, torrefaction, flash pyrolysis and dissolution of wood in organic solvents) on gasification [3]. They reported that torrefaction is the beneficial pretreatment for storage of biomass.

Researcher found that the energy consumption, required for size reduction, deceases for torrefied biomass as compared to raw biomass [4]. They reported that the light and severe torrefaction regimes are not favorable for carbon conversions. Torrefied biomass can be used with coal for co-firing [5,6] but gasification technology has better efficiency and environment friendly performance [7].

Researcher reported that torrefied wood can yield more efficient gasification as compared to raw wood in an entrained flow gasifier [8]. In another work, it has been reported that torrefaction aided gasification is like two-stage pyrolysis gasification process [9], with the difference that heat requirements for torrefaction are lower as compared with pyrolysis. Energy requirements for torrefaction can be met from gasifier product gas. Characteristics of torrefied biomass relevant to gasification were studied [7]. They reported that a more porous and higher surface area biomass is achieved because of torrefaction, which is favorable for gasification.

A coal gasification and biomass torrefaction integrated conceptual system was proposed [10]. Researcher described that the torrefaction [11], as pretreatment process, increases calorific value of biomass and hence torrefied biomass is favorable for gasification because efficiency of gasification is strong function of feedstock calorific value.

Behavior of coal blends i.e., coal with torrefied biomass and raw biomass was studied [12]. They reported the accumulation of material in a fluid-bed gasifier when coal coal-raw biomass blend was used. While, with the utilization of torrefied biomass such accumulation was not observed.

Effect of the torrefied beechwood on gasification was studied [13] in an entrained flow reactor. They reported that with utilization of torrefied wood, O/C ratio decreased, and the quantity of syngas increased.

It is reported that torrefied wood gives improved cold gas efficiency in gasification and better quality of syngas as compared with raw wood [14]. Cold gasification efficiency of torrefied bamboo was higher as compared to raw bamboo. Cold gas efficiency of torrefied biomass was comparable with bituminous coal [15]. Torrefied baggase was used for gasification [11]. Torrefied baggase resulted in increased conversion efficiency of gasification.

Performance of gasification for raw and torrefied bamboo biomass was evaluated using thermodynamic analysis [16]. Their results proved that torrefied biomass can give higher production of syngas as compared to raw biomass [16].

From the above discussion, biomass is the potential source of renewable energy but has limited utilization as source of energy due to low energy density. Torrefaction, as pretreatment process, improves the characteristics of biomass. However, the focus of research has been oriented towards torrefaction only. The effects of torrefaction on the gasification process have not been investigated intensively.

To our knowledge, the effect of torrefaction on the performance of wheat straw gasification has not been studied. The objectives of present study were to quantify the impact of torrefaction on gasification and to find the optimum regime of torrefaction for the efficient gasification of wheat straw. In this study, two models of torrefaction: kinetic model and volatile and solid composition models were used to investigate the behaviors of torrefaction. These models of torrefaction helps to observe the similarities between torrefied biomass and coal, further such similarities have not been studied extensively. Then, the torrefaction models were integrated with gasification model to examine the impacts of torrefied biomass on gasification. From the integrated study of torrefaction-gasification process, the optimum temperature regime for torrefaction was found.

# Modeling

#### Kinetic modeling of torrefaction

A well-developed kinetic model, two-step model [8,17-20] was utilized for the modeling and numerical simulation of torrefaction. Figure 1 shows the two-step reaction mechanism of torrefaction. Table 1 shows the Rate constant of torrefaction [21].



**Figure 1:** Two-step torrefaction reaction mechanism. Where, A=Raw biomass, B=Intermediate product, C=Remaining solid, V=Volatile,  $k_i$ = Reaction rate constant.

$$\frac{d[A]}{dt} = -(k_B + k_{V1})[A] \qquad (1)$$

$$\frac{d[B]}{dt} = k_B[A] - (k_C + k_{V2})[B] \qquad (2)$$

$$\frac{d[C]}{dt} = k_C[B] \qquad (3)$$

$$k_j (T) = A_j \exp\left(\frac{E_j}{RT}\right) \qquad (4)$$

For j=A, B, C, V<sub>1</sub> and V<sub>2</sub>

Biomass	<b>k</b> <sub>B</sub>	<b>k</b> <sub>V1</sub>	κ <sub>C</sub>	<b>k</b> <sub>V2</sub>
Wheat	3.5 ×	3.91 ×	4.34 ×	3.48 ×
Straw	10 <sup>4</sup> exp(-7099 9/RT)	10 <sup>10</sup> exp(-13946 0/RT)	10 <sup>3</sup> exp(-76566 /RT)	10 <sup>7</sup> exp(-11862 0/RT)

 Table 1: Rate constant of torrefaction.

#### Volatile and solid composition model

Compositions of the releasing volatiles and remaining solid were calculated with a similar approach developed [20,22]. The model equation is:

$$\frac{dm_x}{dt} = r_{V1}Y_{x,V1} + r_{V2}Y_{x,V2}$$
(5)

Where,

 $\frac{dm_x}{dt}$ : Rate of volatile 'x' production, rv1, rV2=Reaction rates of releasing volatiles, V1/V2, Yx, V1/V2=Compositional coefficients. To calculate the remaining solid mass, following equation was used:

$$\frac{d(m_{S}Y_{j,S})}{dt} = -r_{V1}Y_{j,V1} - r_{V2}Y_{j,V2}$$
(6)

Where,  $m_s$ =Remaining solid mass,  $Y_{j,x}$ =mass fraction of X (A, B, C, V1, V2)

The high heating value of the torrefied biomass was estimated using Eq (7) and the energy densification ratio of torrefied biomass is defined in Eq (8)

$$HHV\left(\frac{MJ}{kg}\right) = 0.349 \times C + 1.1784 \times H + 0.1005 \times S$$
$$- 0.1034 \times O - 0.0151 \times N - 0.0211 \times Ash \qquad (7)$$
$$e = \frac{(HHV of torrified biomass)}{(HHV of raw biomass)} \qquad (8)$$

#### **Gasification model**

Following model has been utilized by the researchers for gasification process [23,24]. It was adopted for the numerical study of torrefied biomass gasification.

Reaction 1: Boudouard Reaction

$$C_{(s)} + CO_{2(g)} \Leftrightarrow 2CO_{(g)} \Delta \mathrm{H}^{\circ}\mathrm{R} = 172.6 \text{ kJ/mol}$$

$$r_{1} = C_{RF}k_{1} \left( x_{co_{2}} - \frac{x_{co}^{2}}{K_{eq,1}} \right)$$
(9)

Reaction 2: Water Gas Reaction

$$C_{(s)} + H_2 O_{(v)} \Leftrightarrow CO_{(g)} + H_{2(g)} \Delta \mathrm{H}^{\circ} \mathrm{R} = 131.4 \, \mathrm{kJ/mol}$$

$$x_{co} \cdot x_{H_2} \qquad (10)$$

$$C_2 = C_{RF} k_2 \left( x_{H_2O} - \frac{k_{CO} + k_{H_2}}{k_{eq,2}} \right)$$
(10)

Reaction 3: Methanation Reaction

$$C_{(s)} + 2H_{2(g)} \Leftrightarrow CH_{4(g)} \Delta H^{\circ}R=-75 \text{ kJ/mol}$$

$$r_{3} = C_{RF} k_{3} \left( x_{H_{2}}^{2} - \frac{x_{CH_{4}}}{K_{eq,3}} \right)$$
(11)

Reaction 4: Steam Reforming Reaction

$$CH_{4(g)} + H_2O_{(v)} \Leftrightarrow CO_{(g)} + 3H_{2(g)} \Delta H^{\circ}R=206.4 \text{ kJ/mol}$$

$$r_4 = C_{RF} k_4 \left( x_{CH_4} \cdot x_{H_2O} - \frac{x_{CO} \cdot x^3_{H_2}}{K_{eq,4}} \right)$$
(12)

Reaction 5: Water Gas Shift Reaction

$$CO_{(g)} + H_2O_{(v)} \Leftrightarrow CO_{2(g)} + H_{2(g)} \Delta H^\circ R=-41.1 \text{ kJ/mol}$$

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 $r_{5} = C_{RF} k_{5} \left( x_{CO} \cdot x_{H_{2}O} - \frac{x_{CO_{2}} \cdot x_{H_{2}}}{\kappa_{eq,5}} \right)$ (13)

Where,

 $r_i$ =rate of reaction i (mol/m<sup>3</sup>/s)

 $x_j$  = mole fraction of j<sup>th</sup> Component

A<sub>i</sub>=frequency factor for reaction i/s<sup>-1</sup>

C<sub>RF</sub>=char reactivity factor

E<sub>i</sub>=activation energy of reaction i (J/mol/K)

R=gas constant (J/mol/K)

k<sub>i</sub>=rate constant for reaction i

K<sub>eq, i=</sub>equilibrium constant for reaction i

T=temperature (K)

#### Equilibrium constant calculations

Equilibrium constant was obtained using Gibbs function change for all the five reactions as:

$$x_{eq, CO_2} = \frac{x_{eq, CO}^2}{K_{eq, 1}}$$
 (14)

And

$$K_{eq, 1} = e^{-\left(\frac{\Delta G_{2}^{\circ}(T)}{RT}\right)} = \\ -\left(2\frac{g^{\circ}CO^{(T)}}{RT} + \frac{g^{\circ}H_{2}^{(T)}}{RT} + \frac{g^{\circ}H_{2}O^{(T)}}{RT} - \frac{g^{\circ}C^{(T)}}{RT}\right)$$
(15)

Similarly, for all the reactions:

$$\begin{aligned} & -\left(\frac{\Delta G_{2}^{\circ}(T)}{RT}\right) \\ & K_{eq,2} = e^{-\left(\frac{g^{\circ}CO(T)}{RT} + \frac{g^{\circ}H_{2}^{(T)}}{RT} + \frac{g^{\circ}H_{2}O^{(T)}}{RT} - \frac{g^{\circ}C(T)}{RT}\right)}{RT}\right) (16) \\ & = e^{-\left(\frac{\Delta G_{3}^{\circ}(T)}{RT}\right)} = e^{-\left(\frac{g^{\circ}CH_{4}^{(T)}}{RT} - 2\frac{g^{\circ}H_{2}^{(T)}}{RT} - \frac{g^{\circ}C(T)}{RT}\right)}{RT}\right) (17) \\ & K_{eq,3} = e^{-\left(\frac{\Delta G_{4}^{\circ}(T)}{RT}\right)} = e^{-\left(\frac{g^{\circ}CH_{4}^{(T)}}{RT} - 2\frac{g^{\circ}H_{2}^{(T)}}{RT} - \frac{g^{\circ}C(T)}{RT}\right)} (17) \\ & K_{eq,4} = e^{-\left(\frac{g^{\circ}CO(T)}{RT} - 3\frac{g^{\circ}H_{2}^{(T)}}{RT} - \frac{g^{\circ}H_{2}O(T)}{RT} - \frac{g^{\circ}CH_{4}^{(T)}}{RT}\right)} (18) \end{aligned}$$

$$\mathbf{K}_{eq,5} = \mathbf{e} \cdot \left( \frac{\Delta G_5^{o(T)}}{RT} \right)_{= e} \cdot \left( \frac{g^{o}_{CO}(T)}{RT} \cdot 3 \frac{g^{o}_{H_2O}(T)}{RT} \cdot \frac{g^{o}_{H_2}(T)}{RT} \cdot \frac{g^{o}_{CO_2}(T)}{RT} \right)$$

(19)

While,

$$\frac{g^{o}_{i}(T)}{RT} = Ag_{i} + Bg_{i}T + Cg_{i}T^{2} + Dg_{i}T^{3} + Eg_{i}T^{4} + \frac{Fg_{i}}{T} + Gg_{i}lnT \quad (20)$$

The data for the constants is given in Table 2 [25].

Species	$\mathbf{A}_{gi}$	$\mathbf{B}_{gi}$	C <sub>gi</sub>	D <sub>gi</sub>	Egi	F <sub>gi</sub>	<b>G</b> gi
со	-3.08	-7.21 × 10 <sup>-4</sup>	9.38 × 10 <sup>-8</sup>	-8.49 × 10 <sup>-12</sup>	1.16 × 10 <sup>-15</sup>	-1.43 × 10 <sup>4</sup>	-3.03
CO <sub>2</sub>	5.41	-1.57 × 10 <sup>-3</sup>	2.13 × 10 <sup>-7</sup>	-1.99 × 10 <sup>-11</sup>	2.78 × 10 <sup>-15</sup>	-4.90 × 10 <sup>4</sup>	-4.45
H <sub>2</sub>	4.35	-3.50 × 10 <sup>-4</sup>	9.39 × 10 <sup>-9</sup>	7.69 × 10 <sup>-13</sup>	-2.64 × 10 <sup>-16</sup>	-8.35 × 10 <sup>2</sup>	-2.99
H <sub>2</sub> O	-4.19	-1.53 × 10 <sup>-1</sup>	1.46 × 10 <sup>-7</sup>	-1.00 × 10 <sup>-11</sup>	1.07 × 10 <sup>-15</sup>	-2.99 × 10 <sup>4</sup>	-2.67
CH <sub>4</sub>	-7.94	-5.12 × 10 <sup>-3</sup>	6.46 × 10 <sup>-7</sup>	-5.65 × 10 <sup>-11</sup>	7.51 × 10 <sup>-15</sup>	-1.01 × 10 <sup>4</sup>	-1.68
Char	10.2	-8.31 × 10 <sup>-4</sup>	1.11 × 10 <sup>-7</sup>	-1.08 × 10 <sup>-11</sup>	1.53 × 10 <sup>-15</sup>	-7.07 × 10 <sup>2</sup>	-1.49

Table 2: Values of the constants for temperature range 1000-5000 K.

Rate constants for the gasification reactions were calculated using Arrhenius Law Equation [4]. Rate constant data for the gasification reactions is given in Table 3 [22,25].

Reaction <sub>i</sub>	A <sub>i</sub> (sec⁻¹)	E <sub>i</sub> (kJ mol⁻¹)
1	3.616 × 10 <sup>1</sup>	77.39
2	15170 × 10 <sup>4</sup>	121.62
3	4.189 × 10 <sup>-3</sup>	19.21
4	7.301 × 10 <sup>-2</sup>	36.15
5	2.824 × 10 <sup>-2</sup>	32.84

**Table 3:** Rate constant for gasification.

It was proposed [26] that the overall process of gasification can be represented by a single reaction as:

 $\begin{array}{l} CH_{\alpha}O_{\beta}+yO_{2}+zN_{2}+wH_{2}O=x1C+x_{2}H_{2}+x_{3}CO+x_{4}H_{2}O\\ +x_{5}CO_{2}+x_{6}CH_{4}+x7N_{2}\ (21) \end{array}$ 

Where,

 $CH_\alpha O_\beta$  is the chemical representation of dry biomass. The subscripts  $\alpha$  and  $\beta$  are determined from ultimate analysis of torrefied biomass.

y, z, w, xi are the molar numbers of various components.

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At t=0, the initial amounts of the species can be defined as: Compositions of CO and  $H_2$  entering in the reduction zone were assumed to be zero, while the amount of nitrogen remains constant. As nitrogen acts as inert and does not take part in the chemical reaction.

x <sub>2</sub> ,0=0,	x <sub>3,0</sub> =0,	x <sub>7</sub> ,0=z	
and			
x <sub>5</sub> ,0+x <sub>1,0</sub> +	+x <sub>6,0</sub> =1		(22)
2 x <sub>4,0</sub> +4x	<sub>6,0</sub> =a+2w		(23)
x <sub>4,0</sub> +2 x <sub>5,</sub>	$_0=2y+\beta+v$	v	(24)

The above-mentioned equations were solved simultaneously by specifying the compositions from the ultimate analysis of the torrefied biomass.

# Simulation strategy

Simultaneous simulations were run on MATLAB\* for torrefactiongasification integrated process of biomass. Firstly, the torrefaction kinetic, volatile and remaining solids models were solved. Then the ultimate analysis of the torrefied biomass was calculated. The ultimate analysis of the torrefied biomass was input to the gasification model. The gasification model was solved, and the results are presented in Results and Discussion. The conditions of the gasification used in this study are given below.

T<sub>Gasification</sub>=1000 K, C<sub>RF</sub>=100 and Residence time in gasifier=10 sec.

#### **Results and Discussion**

Ultimate analysis of torrefied biomass is given in Table 4. Torrefaction removes the low energy content from the biomass leaving behind high energy dense torrefied biomass. Effect of torrefaction conditions on biomass is discussed in following paragraphs.

#### H/C and O/C molar ratios

Figure 2 shows the behavior of torrefaction impacts on the properties of torrefied biomass. Residence time of 30 minutes was selected for different torrefaction temperatures (473-573 K). H/C molar ratio with H/C molar ratio is shown on Van Krevelan diagram in Figure 2. With the increase in severity of torrefaction conditions, the characteristics of torrefied biomass becomes similar to that of coal. It is clear from the Figure 2 that the utilization of raw biomass in co-firing with coal is not feasible due to the high mismatch in characteristics of both. However, torrefied biomass can be used with coal as blend for co-firing due to the similarity with coal.

At higher torrefaction temperature, torrefied biomass becomes similar to that of lignite and sub bituminous coal. It is due to the removal of low energy volatile contents as a result of torrefaction.

# **Energy densification ratio**

Energy densification ratio reflects that how much the torrefied biomass has been enriched with energy. Effect of torrefaction temperature on energy densification is more pronounced as compared with residence time. At the higher torrefaction temperature, torrefaction removes the higher quantities of low energy volatiles. While, with increasing residence time this effect is not much prominent as shown in Figure 3.



Figure 2: H/C vs O/C molar ratio of torrefied biomass.



#### Effect of torrefaction on gasification

Torrefaction as pretreatment for thermochemical conversions has been promising technology. Improved composition of syngas is obtained as result of the torrefaction-gasification integrated process. Effect of different torrefaction conditions on gasification is shown in Table 4.

Torrefaction conditions		Torrefied biomass			Synthesis gas Composition				
t	т	С	н	0	CO <sub>2</sub>	H <sub>2</sub>	со	H <sub>2</sub> O	CH <sub>4</sub>
min	к	DAF wt %			Mole %				
	Raw	47.50	6.40	45.50	9.40	28.82	26.05	6.77	1.58
60	473	48.25	6.40	44.76	9.31	28.93	25.68	6.58	1.51
60	483	48.71	6.41	44.31	9.28	29.02	25.77	6.55	1.51
60	493	49.33	6.42	43.71	9.24	29.13	25.88	6.52	1.50

60	503	50.16	6.45	42.91	9.18	29.28	26.07	6.49	1.50
60	513	51.34	6.49	41.80	9.10	29.48	26.33	6.45	1.50
60	523	53.05	6.57	40.19	8.99	29.75	26.77	6.40	1.50
60	533	55.64	6.69	37.77	8.83	30.15	27.38	6.33	1.51
60	543	59.62	6.87	34.03	8.59	30.71	28.38	6.24	1.52
60	553	65.76	7.11	28.23	8.26	31.54	29.73	6.09	1.52
60	563	74.91	7.39	19.49	7.82	32.66	31.48	5.85	1.51
60	573	87.71	7.67	7.11	7.29	34.07	33.52	5.52	1.48
30	543	57.83	6.62	35.50	8.59	30.71	28.38	6.24	1.52
60	543	59.62	6.87	34.03	8.51	30.82	28.98	6.29	1.56
120	543	61.69	7.15	32.32	8.47	30.89	29.37	6.33	1.59

Table 4: Effect of raw and torrified biomass on syngas composition.

# Syngas composition with torrefaction residence time and temperature

Figure 4 shows that the torrefaction residence time has little effect on gasification. Increasing, the torrefaction residence time, does not improve the quality of syngas. The reason behind this behavior is that with increasing residence time, properties of biomass are not greatly altered i.e., removal of low energy volatile species is little.



As compared to residence time, torrefaction temperature played important role in alternation of biomass characteristics. As it can be seen from the Figure 5 that composition of syngas was improved at higher temperature of torrefaction i.e., above 520 K. Quality of syngas was much better after the temperature of ~ 550 K. High temperatures provide higher heat contents to remove the volatile species from the biomass. A temperature range of 550-560 K is favorable for torrefaction as pretreatment of gasification keeping in view the economics and torrefied biomass characteristics.

A comparison of syngas composition after gasification for the raw and torrefied biomass is shown in Figure 6. Increase of different syngas component as a result of the torrefaction-gasification integrated process was up to 5% for  $H_2$  and 8% CO. Further, the quantity of  $CO_2$  decreased with increasing severity of the torrefaction making torrefaction favorable pretreatment of gasification.







# Conclusion

Numerical simulations of torrefaction-gasification integrated process were performed with the help of well-developed kinetic, volatile/solid composition and gasification models for the wheat straw biomass. Torrefaction-gasification integrated process gave promising results. Torrefaction as a pretreatment process resulted in improved quality syngas. The content of  $H_2$  and CO increased up to 5% and 8% as a result of integrated process as compared to raw biomass. Severity of torrefaction conditions improved the quality of syngas. The effect of torrefaction temperature was pronounced as compared to residence time.

In future, further research on economic analysis of the integrated process is required for practical implementation of the technology. Overall cost-benefit analysis of the integrated process also needs to be addressed.

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