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Fuel

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Full Length Article

Beech wood gasification in a dense and fast internally circulating fluidized bed

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ARTICLE INFO	A B S T R A C T		
Keywords: Biomass gasification Syngas Olivine Gasifier FICFB	This study deals to improve the understanding of biomass thermo-chemical conversion in a dense and fast internally circulating fluidized bed. The experimental rig used in this study ran either as a dense fluidized bed or as a fast internally circulating fluidized bed. The effect of operating parameters, such as bed temperature (ranging from 750 to 850 °C), steam gas velocity (between 3 and 9.5 times the minimum fluidization velocity of bed material), steam to biomass mass ratio (ranging from 0.7 to 5.6 kg.kg^{-1}) and biomass shape (sawdust and pellets) is investigated in both dense and fast internally circulating fluidized bed with olivine as bed material. In addition, the effect of bed material nature (olivine and sand) is also studied. Results indicated that syngas composition is strongly dependent on steam to biomass mass ratio. Moreover, it was found that syngas yield is favored by rising bed temperature and steam to biomass mass ratio while it is disadvantaged when fluidizing gas velocity increases. In addition, a greater syngas yield was obtained with olivine particles, compared to sand particles. The effect of bed material circulation flow rate was also investigated in fast internally circulating fluidized bed. Results showed that an increase in this parameter leads to a lower syngas yield. Finally, the experimental results presented in this paper were compared to ones reported in the literature and a good agreement was found.		

1. Introduction and state of art

High temperature biomass gasification (> 700 °C) is a promising alternative to fossil fuels for the synthesis of highly energetic products via Fischer-Tropsch or methanation processes. One of the most encouraging and advanced technology is Fast Internally Circulating Fluidized Bed (FICFB) [1]. Its principle relies on the circulation of bed material (catalytic or not) which acts as a heat carrier between an endothermic reactor (called gasifier) where biomass steam-gasification in dense fluidized bed produces synthetic gas (syngas) and an exothermic transported bed reactor (called combustor) where combustion of a part of the solid carbonaceous residue of biomass gasification (char) produces heat. Several demonstrators exist or are in construction to validate this technology and take it to an industrial level.

Biomass steam-gasification is a thermochemical conversion occurring at high temperatures with many simultaneous reactions. Fig. 1 presents a simplified diagram which describes biomass transformation in successive steps.

Starting at 300 °C, the thermal decomposition of biomass, called pyrolysis or devolatilisation, occurs in the absence of oxygen and leads to the formation of:

- Non-condensable products (H₂, CO, CO₂, CH₄ and C_sH_t);
- Volatile condensable products (steam, and primary tars in the form of C_nH_mO_q [2]. Beyond 500 °C, primary tars are converted to non-condensable species (H₂, CO, CO₂, CH₄) and secondary tars in the form of C_nH_mO_q or C_nH_m [2–4]. According to Hosoya et al. [5], chemical functional groups like acids, methoxy, propyl, propenyl, aldehydes and acetyl are eliminated, leaving only aromatic compounds such as cresol and phenol, which constitute the main part of secondary tars. Above 700 °C, secondary tars undergo thermal cracking. In addition, tars can react with gaseous atmosphere in the reactor, like H₂O, CO₂ and H₂, by steam-reforming, CO₂-reforming and hydrodeoxygenation, respectively. These reactions lead to non-condensable gases (H₂, CO, CO₂, CH₄) as well as mono and polyaromatics, called tertiary tars, in the form of C_nH_m [2,6–10]):

Tars steam - reforming: $C_nH_mO_q + (n-q)H_2 O = > (m/2 + n-q)H_2 + nCO$ (R1)

- Tars CO_2 reforming: $C_nH_mO_q + (n-q)CO_2 = > (m/2)H_2 + (2n-q)CO$ (R2)
- Tars hydrodeoxygenation: $C_nH_mO_q + (2n-m/2+q)H_2 = > nCH_4 + qH_2O$ (R3)

Tertiary tars are the most refractory tars regarding thermal cracking reactions, which were found to lead to the formation of many alkenes

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https://doi.org/10.1016/j.fuel.2018.09.025

Received 5 December 2017; Received in revised form 25 July 2018; Accepted 6 September 2018 0016-2361/ © 2018 Published by Elsevier Ltd.





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List of symbols			free biomass)		
		$\mathbf{P}_{\mathbf{i}}$	yield of the component i $(Nm^3 \text{ component i. } kg^{-1} dry ash$		
Ar	Archimedes number (–)		free biomass)		
Cv	coefficient of variance (–)	Re _{mf}	Reynolds number at the minimum fluidization point (-)		
D	wood pellets diameter (m)	T _G	gasifier bed temperature (°C)		
d_{10} , d_{50} , d_{90} particle size distribution (m)		t _{exp}	duration of an experiment (h)		
d _{3/2} (or d _p) Sauter mean particle diameter (m)		U	gas velocity in the gasifier $(m.s^{-1})$		
d _{4/3}	volume particle equivalent spherical diameter (m)	Umf	minimum fluidization velocity $(m.s^{-1})$		
d _{char}	char particle diameter (m)	U _{t,char}	char terminal settling velocity $(m.s^{-1})$		
dp	particle diameter (= $d_{3/2}$) (m)	\dot{V}_i	volumetric flow rate, at STP conditions, of component i		
F _{daf,B}	dry ash free biomass feeding rate $(kg.h^{-1})$		$(Nm^3.h^{-1})$		
F _{H2O}	steam mass flow rate (kg. h^{-1})	Vm	molar volume of a gas, at STP conditions ($Nm^3.mol^{-1}$)		
$F_{H2O}/F_{daf,B}$ steam to biomass mass ratio (kg steam. kg ⁻¹ dry ash		\dot{V}_{N2}	volumetric flow rate, at STP conditions, of tracer gas		
	free biomass)		$(Nm^3.h^{-1})$		
Fp	circulating solid mass flow rate $(kg.h^{-1})$	\dot{V}_T	total volumetric flow rate, at STP conditions, of gas at the		
g	acceleration due to gravity $(m.s^{-2})$		reactor outlet $(Nm^3.h^{-1})$		
H_2/CO	ratio between H_2 and CO molar flow rates in the dry	X _c ^G	carbon conversion rate (-)		
	syngas without tracer (-)	$\mathbf{y}_{\mathbf{i}}$	molar fraction (content) of the component i in the dry		
HHV	higher heating value of the biomass (kJ. kg^{-1} dry ash free		syngas without tracer (–)		
	biomass)		Greek symbols		
L	wood pellets length (m)	γ	compressibility factor (%)		
LHV	lower heating value of the biomass (kJ. kg^{-1} dry ash free	ΔF_B	uncertainty of the biomass feeding rate (kg.h $^{-1}$)		
	biomass) or of the syngas (kJ. Nm^{-3} dry syngas without	ΔF_{N2}	uncertainty of the tracer gas volumetric flow rate		
	tracer)		$(Nm^3.h^{-1})$		
MB	biomass molar weight (kg.mol ⁻¹)	ΔH	enthalpy of reaction (kJ.mol $^{-1}$)		
N _c ^{Bed}	amount of carbon retained in the bed as char (mol)	ΔX_{ash}	uncertainty of the ash content in the biomass (%)		
$N_c^{Biomass}$	amount of carbon fed in the gasifier as biomass (mol)	ΔX_H	uncertainty of the moisture content in the biomass (%)		
N _c ^{Cyclone}	amount of carbon elutriated as char and recovered in the	ΔY	uncertainty of a given performance criteria (same unit as		
	cyclone (mol)		the performance criteria)		
	amount of carbon leaving the gasifier as part of the dry	Δy_i	uncertainty of each component molar fraction (%)		
	syngas (mol)	ϵ_{BA}	aerated bulk bed voidage (-)		
\dot{N}_{c}^{Syngas}	instantaneous molar carbon flow rate leaving the gasifier	μ_{f}	fluid viscosity (kg.m ⁻¹ .s ⁻¹)		
	as part of the dry syngas (CO, CO ₂ , CH ₄ , C ₂ H ₂ , C ₂ H ₄ and	ρ_{BA}	aerated bulk density (kg.m ⁻³)		
	C_2H_6) (mol.h ⁻¹)	ρ_{BP}	packed bulk density (kg.m ⁻³)		
N _c ^{Tar}	amount of carbon converted into tars (mol)	$\rho_{\rm f}$	fluid density (kg.m ⁻³)		
$\dot{N}_{daf,B}$	instantaneous molar flow rate of carbon entering the re-	ρ_p	particle apparent density (kg.m $^{-3}$)		
	actor as biomass (mol. h^{-1})	ρ_{s}	particle skeletal density (kg.m $^{-3}$)		
N_i	molar flow rate of component i $(mol.h^{-1})$	χ	Internal voidage (%)		
P_{G}	syngas yield (Nm ³ dry syngas without tracer. kg ⁻¹ dry ash	\aleph_i^C	carbon atom number in the component i (-)		

like C_2H_2 , C_2H_4 and C_2H_6 [11–13]. Besides, tertiary tars can also undergo steam-reforming as well as steam- and hydrodealkylation, leading to the formation of non-condensable species, such as CH_4 , H_2 and CO, and lighter tars. For instance, Taralas et al. [14] proposed overall steam- and hydrodealkylation reactions schemes for toluene:

Toluene hydrodealkylation: $C_7H_8 + H_2 = > C_6H_6 + CH_4 \Delta H(850 \circ C) = -184.4 \text{ kJ. mol}^{-1}$ (R4)

Toluene steam - dealkylation: $C_7H_8 + H_2 O = > C_6H_6 + 2H_2 + CO \Delta H(850 \circ C) = 40.8 \text{ kJ. mol}^{-1}$ (R5)

It must be precised that reactions R(1) and R(5) strongly depend on steam partial pressure.

A solid carbonaceous residue called char. Morin et al. [15] investigated the effect of the temperature on the composition of char produced by fast-pyrolysis in a dense fluidized bed. The used biomasses were beech pellets and beech sticks, with chemical formula in the form of CH_{1.63}O_{0.79} and CH_{1.71}O_{0.76}, respectively.

Higher carbon content and lower hydrogen and oxygen contents were found for increasing temperatures. For instance, for a bed temperature of 850 °C, char is mainly made of carbon with a chemical formula in the form of $CH_{0.09}O_{0.06}$. For these reasons, some authors considered char as pure carbon [16–18]. Above 700 °C, char reacts with steam to produce hydrogen and carbon monoxide according to:

Char steam - gasification: C+ H_2 O= > CO + H_2 \Delta H(850 \circ C) = 135.6 kJ. mol^{-1}

(R6)

Besides, char can react with carbon dioxide according to:

Boudouard reaction: C+ CO₂ < = > 2CO Δ H(850°C) = 169.2 kJ. mol⁻¹

Tars cracking reactions can lead to the formation of a carbonaceous solid, called coke, at the surface of the bed material [19–22]. This solid will either react with steam, in the gasifier, to form CO and H_2 , or burn in the combustor. In addition, these coking reactions can lead to the formation of soot [23–24].

Steam can also react with carbon monoxide according to:

Water - Gas Shift reaction: CO + H₂ O < = > CO₂ + H₂
$$\Delta$$
H(850°C) = -33.6kJ. mol⁻¹
(8)

The required syngas quality constitutes the main technical and economic challenges for its optimal valorization. The H₂ over CO ratio is an important parameter for catalytic applications like Fisher-Tropsch (H₂/CO > 2) or methanation (H₂/CO > 3). The presence of other species such as methane can be an advantage for methanation, but also reduces the yield of Fischer-Tropsch processes. Besides, refractory tars and inorganic compounds, which are systematically formed, have to be removed to very low concentrations before the downstream processes.

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