On a carbon-negative energy production scheme via a quadruple fluidized bed gasifier

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ABSTRACT

To restrain the CO_2 concentration in atmosphere, deployment of the carbon-negative energy generation devices is indispensable on account of the vast net carbon discharge every year. In this work, a novel and promising quadruple fluidized bed (QFB) reactor for the biomass/coal co-gasification is proposed to generate the high-quality syngas whilst separate CO_2 for further sequestration or utilization. To reveal the fundamental rules governing the reactor, a one dimensional model coupling the gas-solid flow hydrodynamics and the chemical reaction kinetics is built to simulate the physicochemical processes in the QFB, and the effects of several pivotal operation parameters on the reactor performance are comprehensively analyzed. It is found that the QFB runs stably under the given condition. The H_2 mole fraction in the dry syngas is higher than 70%, the CO_2 mole fraction in the dry flue gas is around 97%, and the net carbon discharge can be negative when the biomass blending ratio is over 0.5. The methodology and findings in this work are of reference value for the lab, pilot or industrial scale QFB designation and operation, and can be extended to develop similar novel reactors for the carbon-negative energy generation.

1. Introduction

As is widely advocated, global warming should be restricted below 2 °C to prevent the catastrophic anthropogenic interference with the living environment [1]. To meet this target, it is anticipated that about 42% of the cumulative CO_2 emission between 2009 and 2050 should be mitigated [2], which requires the development of the carbon-negative energy technologies that eliminate CO_2 from atmosphere to compensate the emission from sectors where carbon mitigation is costly. Deployment of bioenergy with carbon capture provides an efficient scenario for the carbon dioxide removal (CDR) relying on photosynthesis [3]. Among the viable ways, it is concluded that the biomass steam gasification is one main thermochemical route to obtain bioenergy, and 4 g (without catalyst) and 7 g (with catalyst) H_2 can be produced from 100 g biomass [4]. However, low calorific value, seasonal dependency and inconvenient collection make biomass untenable and uneconomical to completely substitute fossil fuels for power generation [5]. As a compromise, co-gasification of biomass and coal with in-situ carbon capture offers a compelling route for the large-scale clean energy production since coal is naturally abundant and widely distributed [6]. Besides, some mineral matters (K, Na) in biomass ash can serve as

alternative to the alkali and alkaline earth metal (AAEM) based catalysts to enhance the gasification, exerting the synergistic effect [7].

In that most of the gasification processes are endothermic, additional fuel is usually burnt to balance the heat consumption. If air is chosen as the oxidant, large amount of N_2 dilutes the production, which is adverse either to generate the high-quality syngas or to capture CO_2. If the oxy-fuel combustion technology is chosen, a high projected efficiency penalty (8–12.5% points) is imposed due to the intensive power consumption by the cryogenic air separation unit [8], greatly lowering the overall efficiency. In comparison, the chemical looping process (CLP) [9] for the lattice oxygen generation performs inherent advantage in terms of feasibility, economy and compatibility with the state-of-the-art technologies [10], and can therefore be extended to supply oxygen for the combustion and/or gasification processes. With respect to continuous in-situ CDR during the co-gasification, the CaO/$CaCO_3$ based calcium looping (CaL) offers a viable and economical [11] alternative to the chemical solvent scrubbing approach, and has been widely tested in bench or pilot scale plants worldwide [12].

With all the aforementioned technologies well developed, the only issue to be solved to realize the biomass/coal co-gasification based carbon-negative energy production scenario is the development of a

Nomenclature

a	decay constant
A_b	bubble phase cross-sectional area, [m^2]
A_d	ash content in dried basis, [–]
A_i	pre-exponential factor [1/s]
Ar	Archimedes number
A_t	bed cross-sectional area, [m^2]
C_b	concentration in bubble, [mole/m^3]
C_e	concentration in emulsion, [mole/m^3]
C_j	concentration of species j, [kmole/m^3]
$C_{p,p}$	particle specific heat, [J/(kg·k)]
d_p	particle diameter, [m]
D_B	bubble diameter, [m]
D_{B0}	initial bubble diameter, [m]
D_{BM}	maximum bubble diameter, [m]
D_j	diffusion coefficient, [m^2/s]
D_t	column diameter, [m]
E_i	activation energy [kJ/mol/K]
f_i	mole fraction of species i
F_b	molar flow rate in bubble, [mole/s]
F_{biom}	biomass mass flow rate [kg/s]
F_c	circulating flux in combustor [kg/m^2/h]
F_{CaO}	CaO mass flow rate [kg/s]
F_{coal}	coal mass flow rate [kg/s]
F_{dc}	discharged carbon mass flow rate [kg/s]
F_e	molar flow rate in emulsion, [mole/s]
F_f	feedstock mass flow rate [kg/h]
F_r	circulating flux in reductor [kg/m^2/h]
F_s	steam flow rate, [kg/h]
g	acceleration of gravity, [m/s^2]
h_{be}	bubble-to-emulsion heat transfer coefficient, [W/(m^2·K)]
h_{gp}	gas-to-particle heat transfer coefficient, [W/m^2/K]
H	total enthalpy flow , [kJ/s]
ΔH	enthalpy generation, [J/mole]
k	reaction rate constant
k_g	gas thermal conductivity, [W/(m·K)]
k_s	solid thermal conductivity, [W/(m·K)]
K_{bd}	bubble-to-dense phase mass transfer coefficient, [1/s]
L_d	dense bed height, [m]
L_f	freeboard height, [m]
M_{ar}	moisture in as-received basis, [–]
M_s	mass of bed material, [kg]
$M_{s,i}$	mass of solids in i_{th} stage, [kg]
n	total number of species, [–]
n_d	number of orifices in distributor, [–]
p	operation pressure, [Pa]
P_j	partial pressure of species j, [bar/atm]
Pr_p	Prandtl number, [–]
$r_{(i),j}$	consumption rate of species j in stage i, [kmole/(m^3·s)]
r_{blend}	net increment of r_{coal} [1/s]
r_{coal}	conversion rates of coal char [1/s]
r_{mix}	conversion rates of blended char [1/s]
R_b	biomass blending ratio
R_c	char conversion
R_{cal}	Ca/C molar ratio
Re_p	Reynolds number, [–]
R_g	gas constant, 8.3145, [J/K]
R_{H2O}	steam conversion, [–]
R_{sc}	steam-to-carbon ratio, [–]
S_{cf}	sand circulation flux, [kg/(m^2·s)]
T_b	temperature in bubble, [K]
T_e	temperature in emulsion, [K]
T_g	initial gasification temperature [°C]
T_o	initial oxidation temperature [°C]
U_0	superficial velocity, [m/s]
U_{br}	bubble rise velocity, [m/s]
U_{mb}	minimum bubbling velocity, [m/s]
U_{mf}	minimum fluidization velocity, [m/s]
V_d	volatile in dry basis, [–]
V_b	volume of bubble phase, [m^3]
V_e	volume of emulsion phase, [m^3]
x	conversion [%]
Y_{biom}	yield from biomass [%]
Y_{coal}	yield from coal [%]
Y_{tv}	volatile fraction by calculation [%]
Y_v	volatile fraction by proximate analysis [%]
z	length of PFR, [m]
z_f	height above distributor, [m]

Greek symbols

δ_b	bubble phase volume fraction, [–]
ε_b	bubble phase voidage, [–]
ε_{mf}	emulsion voidage at minimum fluidization, 0.4, [–]
μ_g	gas viscosity, [kg/(m·s)]
ρ_g	gas density, [kg/m^3]
ρ_p	particle density, [kg/m^3]
τ	residence time of particle in bubble, [s]
ϕ_d	dense phase solids volume fraction, [–]
ϕ^*	saturation carrying capacity, [–]
ϕ_f	freeboard solids volume fraction, [–]
ψ_{char}	one minus char conversion, [–]
ψ_i	structure parameter

Abbreviations

AAEM	alkali and alkaline metal
BAM	bubble assemblage model
BBM	black box model
BFB	bubbling fluidized bed
CaL	calcium looping
CBM	countercurrent backmixing model
CDR	carbon dioxide removal
CFDM	computational fluid dynamics model
CLP	chemical looping process
CSTR	continuous stirred tank reactor
DFB	dual fluidized bed
DHM	Davidson-Harrison model
FFB	fast fluidized bed
KLM	Kunii-Levenspiel model
LHV	low heating value
PFR	plug flow reactor
PS	pine sawdust
SC	Shinhwa coal
WGS	water-gas shift

proper reactor. Among all the CLP oriented reactors involving the rotating bed, the alternating fixed bed, the moving bed and the fluidized bed, the most suitable reactor for the large scale redox process is probably the dual fluidized bed (DFB), in which the particulate oxygen carrier is reduced to release oxygen in one side while oxidized to absorb oxygen in the other, with the dual beds serially coupled by the cyclic metal oxides [13]. As the core of CLP, the oxygen carriers now exhibit great diversity after decades of years development. Since different oxygen carriers behave differently due to distinct molecular structures, selection of the proper oxygen carrier then becomes essential, and the