MODELLING OF THE FLUIDIZED BED SORPTION ENHANCED STEAM GASIFICATION CONSIDERING PARTIAL LOAD OPERATION

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Abstract - SER (Sorption Enhanced Reforming) steam gasification of biomass in a DFB (Dual Fluidized Bed) reactor system is seen as a technology with great potential to extend the share of renewable energies by compensating fluctuating power production from wind and solar sources and by allowing the production of transportation fuels and chemicals from biogenic feedstock material in a poly-generation approach (e.g. methanation, methanol, Fischer-Tropsch or dimethyl ether synthesis). Using calcium oxide as bed inventory, CO₂ formed during gasification can be captured by the reversible carbonation reaction (CaO + CO₂ \leftrightarrow CaCO₃), yielding a H₂ rich syngas. The carbonation reaction is temperature dependent and depending on the selected gasification temperature, hydrogen concentrations of up to 75 vol%db can be reached, while the amount of produced synthesis gas depends on the plant load (supplied biomass feed). The possible load range in a DFB gasifier is mainly determined by fluid dynamics in the bubbling fluidized bed reactor. The lower limit is given through a minimum fluidization in the reactor and the upper limit is defined by the transition to a circulating fluidization mode. To investigate the gasification performance, a stationary, one dimensional model for a 200 kW_{th} fluidized bed gasification system was developed and applied for variations in gasification temperatures and plant loads. Concentrations of gas and solids were calculated over the reactor height and the fluidization regime was determined by velocity profiles.

INTRODUCTION

With increasing installed capacity of intermittent renewable power production systems, such as photovoltaics and wind power, specific system-related measures are required to maintain a reliable operation of the the power system (e.g. increased flexibility of conventional power plants, installation of energy storage systems) (Neubarth, 2011). In this context, the gasification of biomass is a promising and flexible technology since the generated synthesis gas can either be used directly for generating electricity (gas turbine, gas engine) or for synthesis of liquid or gaseous energy carriers that can be stored. An example for this approach is the methanation and energy storage in the natural gas network (Substitute Natural Gas, SNG). Besides the option of switching the operation mode between power generation and energy carrier production for energy storage, also a part load operation of the gasifier can be a measure to flexibly compensate the power grid's load requirements. Therefore, in the power generation mode, a good partial load operational behaviour is of crucial importance.

GASIFICATION PROCESS DESCRIPTION

This paper considers the SER gasification process as illustrated in Fig. 1.



Fig. 1. Schematic of the sorption enhanced reforming steam gasification process

This process is realized in two interconnected fluidized bed reactors: A steam blown gasification reactor (gasifier) and an air blown combustion reactor (regenerator). By using steam as gasification medium and fluidization agent, a nitrogen free syngas with a relatively high LHV can be produced. The necessary heat for

the endothermic pyrolysis and gasification reactions is provided by circulating bed material (CaO) that is heated up to 850-930 °C in the circulating fluidized bed regenerator. The hot CaO particles enter the bubbling fluidized bed gasifier where they react via the exothermic carbonation reaction (CaO + CO₂ \rightarrow CaCO₃) to form calcium carbonate (CaCO₃). At the bottom of the bubbling bed, the CaCO₃ and unconverted char from the biomass gasification exit the gasifier via a loop seal. The extracted solids stream (600-850 °C) is transferred to the regenerator where the bed material is heated up again by combustion of the char (and additional biomass) and CaCO₃ is calcined (CaCO₃ \rightarrow CaO + CO₂) releasing the CO₂ captured during gasification. As the carbonation and calcination are equilibrium reactions, the temperature affects the amount and compositions of products/educts in both reactors and hence, the gasification temperature is an important parameter to adjust the composition of syngas for different applications. The CO₂ capture in the gasification reactor also impacts the equilibrium of the water-gas shift reaction allowing to produce syngas with hydrogen concentrations of up to 75 vol%_{db}.

MODEL DESCRIPTION

A stationary, one dimensional model for a 200 kW_{th} bubbling fluidized bed gasification system was developed to determine the impact of the gasification temperature and different plant loads on the system performance. The model considers the bubble and suspension phases in the fluidized bed, respective models for material transport and specific reaction kinetics for the reactions in SER steam gasification. Gas and solids concentrations are calculated over the reactor height and the fluidization regime is determined by velocity profiles. The model was verified on the basis of experimental data and can be used for the design of experiments.

Chemical reaction system

In Table 1 the chemical reactions used in the model to describe the sorption enhanced steam gasification are listed. Additionally, literature references providing respective kinetic data and the phase in which each reaction can takes place (d: suspension phase, b: bubble phase, f: freeboard) are mentioned.

Table 1: Chemical reactions to describe sorption enhanced steam gasification, respective references providing kinetic data and the phases that are considered for each reaction (d: suspension phase, b: bubble phase, f: freeboard)

Reaction	Reaction equation			Phase
Drying	BM _{a.r.}	\rightarrow	$BM_{\rm wf} + H_2O$	d
Primary pyrolysis (Boroson et al., 1989), (Di Blasi, 2004)	$BM_{\rm wf}$	\rightarrow	$\begin{array}{l} C+Tar+Ash+H_2O+CO_2\\ +CO+CH_4+H_2 \end{array}$	d
Secondary pyrolysis (Boroson et al., 1989), (Di Blasi, 2004)	Tar	\rightarrow	$\mathrm{CO}_2 + \mathrm{CO} + \mathrm{CH}_4 + \mathrm{H}_2$	d, b, f
Heterog. water-gas reaction (Di Blasi, 2009)	$\mathrm{C}+\mathrm{H_2O}$	\rightarrow	$\rm CO + H_2$	d
Boudouard reaction (Kramb et al., 2014)	$\mathrm{C}+\mathrm{CO}_2$	\leftrightarrow	2 CO	d
Heterog. methane synthesis (Di Blasi, 2004)	$\mathrm{C}+2~\mathrm{H_2}$	\rightarrow	CH ₄	d
Methane reforming (Tepper, 2005)	$\mathrm{CH}_4 + \mathrm{H}_2\mathrm{O}$	\rightarrow	$\rm CO+3~H_2$	d
Water-gas shift reaction (Di Blasi, 2004)	$\rm CO + H_2O$	\leftrightarrow	$\mathrm{CO}_2 + \mathrm{H}_2$	d, b, f
Carbonation (Charitos et al., 2011)	$CaO + CO_2$	\leftrightarrow	CaCO ₃	d

The elemental and proximate analysis of the biomass (wood pellets) that was used for experiments and is considered in simulations is shown in Table 2.

Table 2: Elemental	l analysis	of wood	pellets
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C	Н	0	Ν	S	Ash	H_2O	LHV
·		wt‰ _{waf}			wt‰ _{wf}	wt‰ _{a.r.}	MJ/kg
49.0	6.97	44.1	n.d.*	n.d.*	0.350	9.70	15.6

*not detectable

Description of fluid dynamics

A two-phase model according to (Werther, 1972) and (Hilligardt, 1987) was used in this work, to describe the fluid dynamics of CaO/CaCO₃ particles with a diameter of 350 μ m. The model distinguishes between a porous suspension phase and a solid-free bubble phase. The calculation of volume fractions and gas velocities of the phases as well as the material exchange between the phases is based on empirical equations according to (Hilligardt, 1987), with an extension for continuity equations for each phase. This is necessary to account for specifics of the SER gasification reaction and the reactor system, i.e. a secondary steam and biomass inlet above the primary steam inlet, source and sink terms due to gasification reactions and a changing reactor cross section over the height in the fluidized bed area (see Fig. 2, left side for details of the reactor geometry). In cold model studies corresponding to the 200 kW_{th} fluidized bed gasifier, Bidwe et al. (2014) observed a complex velocity field including different back mixing zones in the fluidized bed. The movement of the solids in the bubbling fluidized bed is mainly driven by ascending gas bubbles, which displace solids or carry solids in their wake and drift. The solids mass flow, induced by wake and drift are calculated according to equations by Tepper (2005).



Fig. 2. Left side: Illustration of the geometry of the 200 kW_{th} gasification reactor and the observed velocity field of solids and bubbles from cold model studies (modified based on Bidwe et al. (2014)); Right side: Fluid dynamic model and balance equations

Discretization and balance equations

For the one-dimensional description of the gasification reactor, the volume of the reactor was discretized to finite cells (number n) in axial direction. For each cell, mass and energy balances for the suspension phase (P = d) and the bubble phase (P = b) were considered (Fig. 2, right side). In the gasification reactor, beside the upward directed transport of solids from wake and drift, there is also a superimposed downward directed transport due to the solids extraction at the bottom of the reactor and due to the changing cross section of the reactor. This leads to a strong back mixing in the bubbling bed of the gasification reactor. To consider these

effects, the model includes a complete transport of the drift mass flow to the next upper cell with an ideal mixing, while the wake is only able to carry five percent into the calculation cell while the rest remains attached to the bubble. This is considered in the balance equations by a cell-bypass and can be set up by the parameter alpha (Fig. 2, right side; Eq. (4)). It enables a precise modeling of the material and thermal mixing characteristic of the considered fluidized bed facility. The gas flows are assumed to be not back mixed.

To balance the energy within a cell, an ideal mixing of all enthalpy flows incoming and outgoing via gas and solid components is assumed. An exception is the wake mass flow in the cell bypass, which is only completely mixed with the remaining fluidized bed at the top computational cell in the transition to the freeboard. The energy balance additionally takes into account reaction enthalpies, feed streams entering along the reactor height (secondary steam, regenerated bed material), and evaporation enthalpy of the fuel moisture and heat losses over the reactor jacket surface.

VERIFICATION OF THE MODEL

To verify the model, experimental results from a corresponding 200 kW_{th} pilot test facility and also from a 20 kW_{th} electrically heated test facility were compared to the simulation results. From Fig. 3 it can be seen that the model is able to describe the measured gas concentrations as well as the yield of the syngas in a wide temperature range. Looking at the course of the simulated gas yield, one can see a good agreement with the measured values.



Fig. 3. Gas concentrations and yield of syngas for different gasification temperatures: Comparison of simulation results and experimental data from a 200 kW_{th} and a 20 kW_{th} gasification test facility (experimental data by Poboss et al., (2013))

Below gasification temperatures of 750 °C, gas concentrations are highly affected by the carbonation reaction that cases the in-situ capture of CO_2 from the syngas. In consequence, the depletion of CO_2 shifts the equilibrium of the water gas shift reaction towards hydrogen concentrations of up to 75 vol%_{db}. For gasification temperatures above 750 °C, the equilibrium of the carbonation reaction is on the side of calcium oxide and hence CO_2 cannot be captured. This leads to significantly lower hydrogen concentrations in the syngas.

In addition to syngas composition, also the calculated temperature profile over the reactor height was compared to experimental data from the 200 k W_{th} fluidized bed gasifier (Fig. 4).



Fig. 4. Simulated temperature profile over the reactor height of the 200 k W_{th} fluidized bed gasifier: Comparison to experimental data by Hawthorne et al. (2012)

The simulation model only considers the fluidized bed region of the reactor system without the loop seal at the bottom which is required in the DFB configuration to maintain a constant solid mass flow to the regenerator. The model starts with the first computational cell at the level of the primary steam inlet (sparger at 0 m). A relatively constant temperature profile can be observed both in the measured values and in the calculated temperature profile up to approximately 1.1 m, which represents the height of the bubbling fluidized bed. This suggests a good mixing of solids in the fluidized bed. The maximum temperature is reached in the freeboard, at the inlet of the hot solids transferred from the regenerator. Downstream, the temperature in the freeboard decreases with increasing reactor height to around 500 °C at the reactor outlet due to heat losses. For the selected operational parameter (steam-to-carbon ratio S/C = 2.2 mol/mol, $\dot{M}_{BM,a.r.} = 30$ kg/h, see Hawthorne et al. (2012)), the model is able to predict the profile of measured temperatures.

SIMULATIONS OF PARTIAL LOAD OPERATION

Following the verification of the model by comparison of simulated and experimental data, it was applied to investigate the partial load operational behavior of the SER process for gasification temperatures of 650, 710 und 750 °C. Based on the syngas concentrations (see Fig. 3), an operational point of around 650 °C with its high hydrogen concentrations is well suited for production of hydrogen while 750 °C with higher syngas yields will be very good for a power generation application. For synthesis of fuels etc., temperatures in between would be ideal. For this purpose, the syngas is required to provide a stoichiometric composition to enable a full conversion to the desired product. For example, a temperature of 710 °C would be favourable for SNG synthesis, according to the methanation reactions ($3H_2 + CO \leftrightarrow CH_4 + H_2O$, $4H_2 + CO_2 \leftrightarrow CH_4 + 2H_2O$).

For the three different gasification temperatures, a parameter study for different plant loads (i.e. different biomass feed rates) was carried out to receive profiles of superficial gas velocities over the reactor height (Fig. 5) using the model previously introduced. In the velocity profiles up to 0.35 m, the influence of the conical reactor geometry leading to a decreasing gas velocity and of the biomass and secondary steam feeds increasing the gas velocity can be recognized (compare reactor geometry in Fig. 2). Additionally, Fig. 5 also provides information on the calculated height of the bubbling fluidized bed (h_{bed}), which slightly increases at higher temperatures and considerably increases at higher plant loads. Above the bed (i.e. $h = h_{bed}$), the gas velocities increase up to the inlet level of hot solids from the regenerator and then decrease due to heat losses.

The possible load range for the SER steam gasification can be defined by requirements of the bubbling fluidized bed according the fluid dynamics. The lower limit is given through a minimum fluidization to guarantee a homogeneous mixing of solid particles in the bed and to maintain a sufficient solid circulation between gasifier and regenerator. The upper limit is mostly defined by the transition to a turbulent/circulating fluidization regime in order to avoid a high entrainment and discharge of solids from the reactor. This limit is experienced at the top of the bubbling fluidized bed at the transition to the freeboard and from Fig. 5 corresponding superficial gas velocities can be extracted to identify the fluidization regime for different temperatures and loads. It can be recognized that gasifier operation at lower gasification temperatures and with low loads may be limited by an insufficient mixing in the bubbling bed. In contrast,

operation conditions at higher temperatures and high loads will be limited by an increased discharge of bed inventory due to high superficial gas velocities at the top of the bubbling fluidized bed.



Fig. 5. Influence of biomass (BM) feed rate and gasification temperature on reactor fluidization (superficial gas velocity u) and calculated height of fluidized bed (h_{bed})

CONCLUSION

A stationary, one dimensional model for a 200 kW_{th} bubbling fluidized bed sorption enhanced steam biomass gasification system was developed, considering the fluid dynamics and reactions kinetics for this process. The model was compared to experimental data of a 200 kW_{th} and a 20 kW_{th} gasification test facility and showed a good agreement with this data. Based on the determined process characteristic, operating temperatures for hydrogen production, SNG synthesis and power generation were identified. In a further step, the influence of different plant loads on the superficial gas velocity along the reactor height was investigated by simulations. It could be recognized that an operation at low gasification temperatures and low plant loads may be limited by an insufficient mixing in the fluidized bed, while high gasification temperatures and high loads will lead to a higher rate of entrainment and discharge of the bed inventory from the bubbling fluidized bed.

NOTATION

α	fraction of cell bypass	Ń	mass flow, kg/s
a.r.	as received	MW	Molar weight, kg/kmol
b	index for bubble phase	n	number of cell
bed	fluidized bed	ν	stoichiometric coefficient, -
BM	biomass	Р	general phase index
d	index for dense phase	R	reaction rate, mol/s
db	dry basis	r	volume specific reaction rate, mol/m3/s
DFB	Dual Fluidized Bed	S/C	steam-to-carbon ratio, mol _{H2O} /mol _C
dh	height of calculation cell, m	SER	Sorption Enhanced Reforming
dr	drift	SNG	Substitute Natural Gas
Е	Exchange of mass	STP	Standard Temperature Pressure
f	index for freeboard	th	thermal
h	height, m	u	velocity, m/s
het	heterogeneous chemical reactions	W	wake
i	reaction number	\mathbf{W}_1	mixed part of wake
in	inflow of mass	W ₂	not mixed part of wake
j	gas component	waf	water ash free
k	solids component	wf	water free
LHV	Lower Calorific Value, MJ/kg		

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