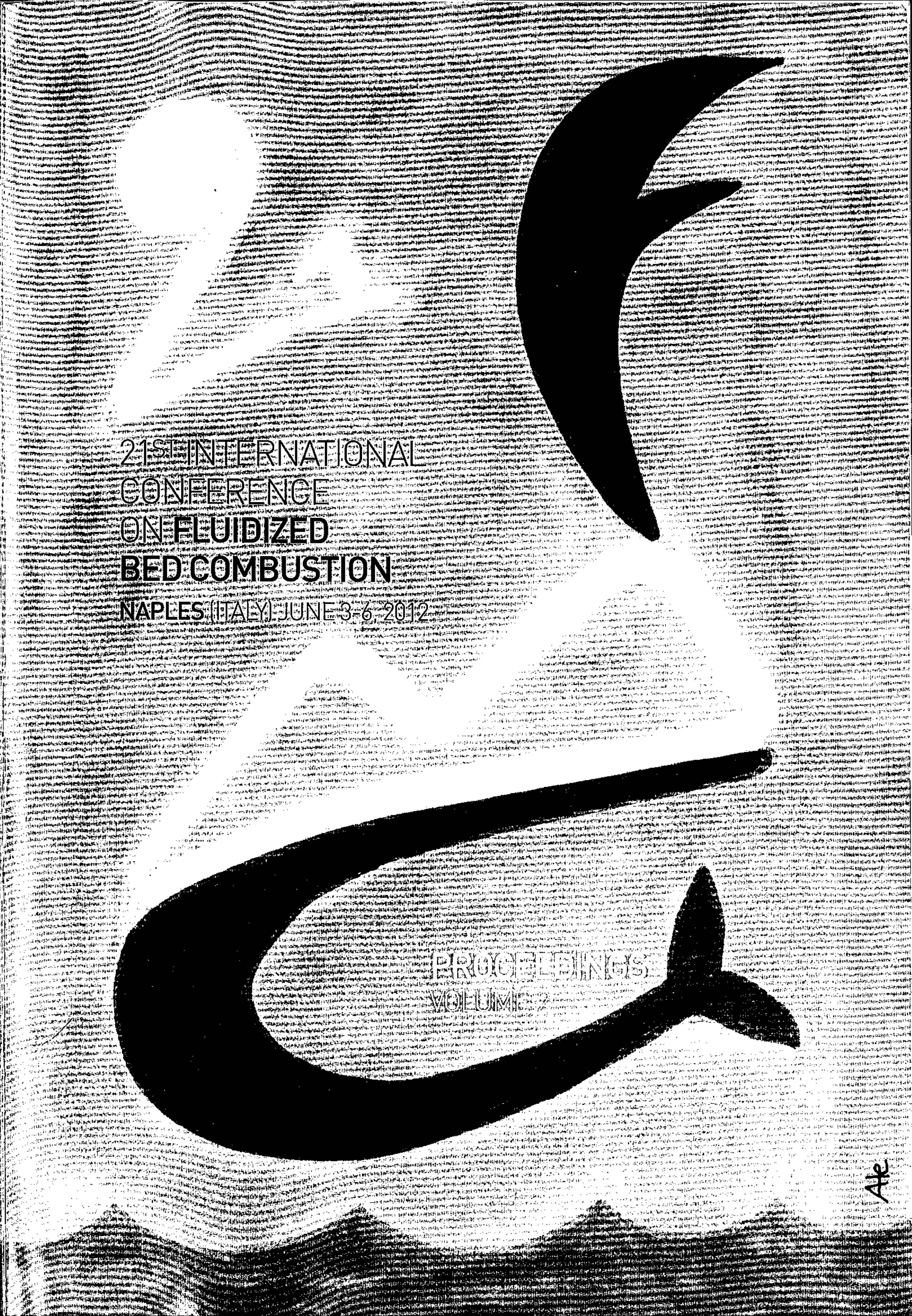

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A THERMODYNAMIK INVESTIGATION OF DUAL FLUIDIZED BED BIOMASS GASIFICATION WITH SORPTION ENHANCED REFORMING

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Abstract: A mass and energy balance model of a dual fluidized bed system for steam gasification of solid biomass is presented. The process model includes a description of solid streams and thermodynamic aspects of solids CO_2 transport capacity for selective CO_2 transport from the gasification zone to the combustion zone through a $CaO/CaCO_3$ system (sorption enhanced reforming). The model is able to verify experimental data with high hydrogen content in the product gas if adequate temperatures are operated in gasification and combustion zone. Parameter variations using the model show that there is a critical trade-off between CO_2 transport potential and global heat integration potential depending on the solids circulation rate operated. Based on typical CO_2 transport capacity values for calcium based sorbents dependent on cycle number, an optimum bed material renewal rate can be found. If a high H_2 content in the product gas is the main aim, high renewal rates and lower energy efficiency must be taken into account. Such a configuration may be suitable in combination with industrial units processing large amounts of limestone or lime.

Keywords: gasification, sorption enhanced reforming, modeling, biomass, thermodynamic limits

INTRODUCTION

Biomass steam gasification in a dual fluidized bed allows conversion of solid fuel into a medium calorific gaseous fuel mainly consisting of hydrogen, carbon monoxide, methane, carbon dioxide and water. The subsequent production of synthesis products like synthetic natural gas, liquid hydrocarbon fuels or even pure hydrogen, is currently investigated by several research groups around the world. So far demonstrated dual fluidized bed gasification units mainly used olivine as bed material and focused on the production of heat and electricity in gas engines (Hofbauer 2007). The application of limestone as bed material instead of olivine has been well known as a part of the sorption enhanced reforming process for many years and promises an improvement of the achievable gas composition by selective elimination of CO_2 from the product gas mixture (Koppatz 2008). But the achievable performance of limestone as bed material in a dual fluidized bed for sorption enhanced reforming is limited. In addition limiting key operation parameters like **reaction temperatures, solids circulation rate, bed material renewal rate** and **decay of CO_2-transport capacity** are significantly influencing the **cold gas efficiency.**

Discussions about the achievable efficiency of sorption enhanced reforming often lead to a situation where thermodynamic limitations are not adequately considered. The main limitation of the process results from the dilemma of suitable solids circulation rate. If the solids circulation rate is low, the sorbent capacity limits the amount of CO_2 transported from the gasifier to the combustor. If the solids circulation rate is high and provides enough transport capacity, the necessary temperature difference between gasifier (carbonator) and combustior (calcinator) cannot be kept. A precise description of the thermodynamics allows to direct specific experimental investigations based on the presented results. Activities of research and development already illustrated main limiting factors for sorption enhanced reforming in the past. Abanades (2002) showed an approach for the calculation of maximum CO_2 capture efficiency at the sorption enhanced reforming process. In a further step Abanades et al. (2003) demonstrated conversion limits in the reaction of CO_2 with lime by experimental investigations. Followed by Grasa et al. (2006) who illustrated the behavior of the CO_2 capture capacity of CaO in long series of sorption enhanced reforming by experimental investigations. The outcome of these works is a good basis for the assessment of achievable performance of CaO particles within the sorption enhanced reforming process. Limitations for the practical application of sorption enhanced reforming in a dual fluidized bed biomass gasification system were investigated by Soukup et al. (2009). Pfeifer et al. (2009) showed the results of experimental investigations with a 100 kW gasifier operating sorption enhanced reforming. Furthermore, Koppatz et al. (2009) showed the results of a large scale experiment with an 8 MW biomass gasification plant in Güssing, Austria. While the results of mentioned research activities showed promising aspects (Koppatz et al. 2009), the demonstration of large scale gasification systems using the sorption enhanced reforming still implies risks. Major risks for the process are loss of CO_2 transport capacity and high bed material consumption due to attrition (Soukup 2009). Both risks have a significant impact on the achievable process performance which is often underestimated. The question which overall plant performance could be theoretically achieved, has not been satisfactorily answered so far. The present work tries to find an answer to this question. Based on the earlier work of Pröll et al. (2008) the main objective of the present work is to illustrate the theoretical limitations of gasification supported by sorption enhanced reforming. Therefore, the thermodynamic limitations are investigated with a process modeling approach reflecting previous published experimental data. Existing limitations are investigated with a special focus on the theoretical achievable performance of lime stone particles. A thermodynamic variation of the performance of used solid particles is made while reaction temperatures and a constant describing the distance to water-gas-shift equilibrium are set as fixed values. Achieved findings and results should reveal fundamental relationships influencing the achievable energetic efficiency and enable a concise prediction of main relationships for the design of experimental facilities.

METHODOLOGY AND THEORETICAL BACKGROUND

The process simulation software IPSEpro is used for the calculation of the thermodynamic limits of gasification combined with sorption enhanced reforming. IPSEpro calculates mass- and energy balances for a created simulation model. **Fig. 1** (a) shows a schematic description of the investigated process configuration. This process configuration is modeled for a reference case. The reference case, shown in **Table 1**, reflects key operation parameters and should represent a starting point for variations investigating the theoretical limit for the process performance. Based on thermodynamic equilibrium calculations and achievable solid particles performance, the reachable energy efficiency is investigated. Data for solid particles performance are taken from experimental results of Grasa et al. (2006). Experimental results implemented in a validated simulation model should provide solid figures about the maximum achievable process performance.

The pictured reference case is the starting point for variations reflecting key operation parameters relevant for the design of experimental facilities as well as future industrial facilities. Key operation parameters are reaction temperatures, the solids circulation rate, the bed material renewal rate and the decay of CO_2-transport capacity. As already mentioned in the introduction, carried out variations are made with a special focus on the thermodynamic performance of used solid particles. During the variations **reaction temperatures** are defined as **constant values** which enhance the carbonation and calcination reaction. Lower gasification temperatures favor selective transport of CO_2 from the gasifier to the combustor due to the equilibrium of the carbonation reaction. Additionally, this leads to a higher H_2

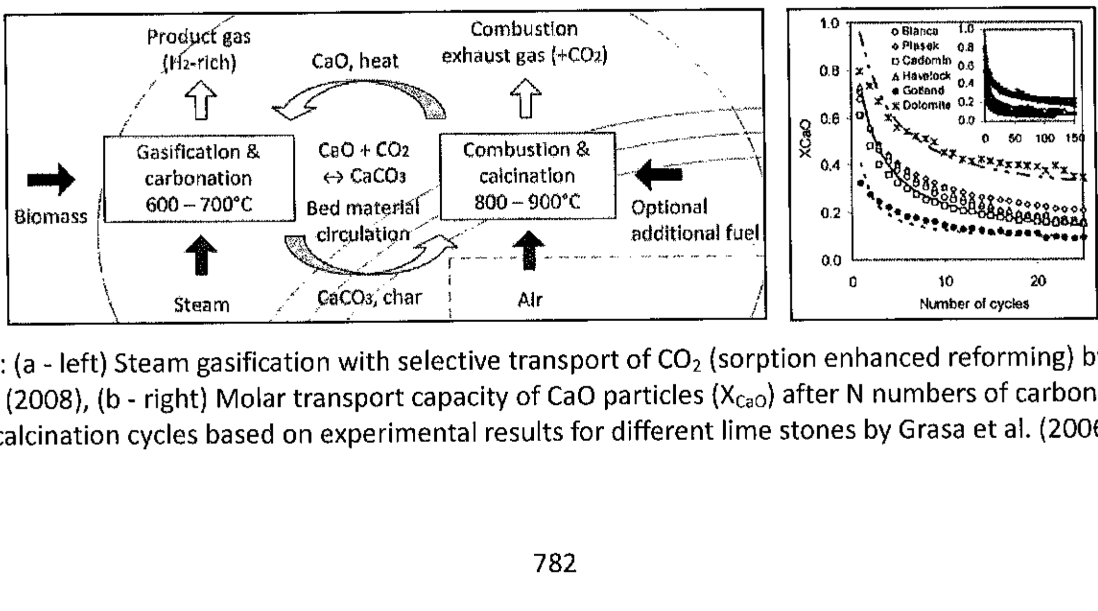

Fig. 1: (a - left) Steam gasification with selective transport of CO_2 (sorption enhanced reforming) by Pröll et al. (2008), (b - right) Molar transport capacity of CaO particles (X_{CaO}) after N numbers of carbonation-calcination cycles based on experimental results for different lime stones by Grasa et al. (2006)

Table 1: Reference case for variations within the created IPSEpro simulation model

Constant parameters during simulation runs	Symbol	Unit	Reference case value
Thermal fuel power (LHV of biomass, water content 20%)	P_{th}	kW	10 000
Optional additional fuel power	P_A	kW	0
Heat loss of overall system	Q_{loss}	kW	225
Gasification temperature	T_G	°C	645
Combustion temperature	T_R	°C	900
Air ratio for combustion	λ_R	-	1.1
Share of CO in relation to CO_2 in combustor exhaust gas	σ_{CO}	mol/mol	0
Fluidization air temperature	$T_{fluid,R}$	°C	450
Fluidization steam temperature	$T_{fluid,G}$	°C	450
Fluidization mass flow gasifier (steam)	$\dot{m}_{fluid,G}$	kg/h	600
Share of CaO in bed material entering the gasifier from combustor	$\omega_{CaO,bed,G,in}$	kg_{CaO}/kg_{bed}	0.85
Share of $CaCO_3$ in bed material entering the gasifier from combustor	$\omega_{CaCO3,bed,G,in}$	kg_{CaCO3}/kg_{bed}	0.15
Total amount of bed material within the system	$m_{inventory}$	kg	3 000
Amount of bed material within gasifier	$m_{gasifier}$	kg	1 500
Cross-sectional area of gasifier	A_G	m^2	2.95
Logarithmic distance to equilibrium (water-gas-shift reaction, gasifier)	$\delta_{Eq.,WGS\text{-}shift}$	-	0.0

content in the product gas stream. But at the same time, lower gasification temperatures reduce a favorable gasification process constraining char gasification reactions due to short residence times of solid fuel particles at low gasification temperatures (Florin 2008). Therefore set values for reaction temperatures are based on experimental results which demonstrated a stable operation of gasification by sorption enhanced reforming (Koppatz et al. 2009). Lower combustion temperatures would increase the cold gas efficiency while higher combustion temperatures favor the calcination reaction which is especially important when low residence times occur or oxyfuel combustion is applied. The combustion temperature (900°C) has been set on a quite high level and favors calcination reactions at low residence times. Solids circulation rate is investigated by the calculation of the introduced parameter **calcium circulation rate**. The calcium circulation rate is introduced with respect to the performance of solid CaO particles. Therefore, following equation is used:

$$(1) \qquad \delta_{CaO} = \frac{\dot{m}_{bed,G,in} \cdot \left(\frac{\omega_{CaO,bed,G,in}}{M_{CaO}} + \frac{\omega_{CaCO_3,bed,G,in}}{M_{CaCO3}}\right)}{m_{inventory} \cdot \left(\frac{\omega_{CaO,inventory}}{M_{CaO}} + \frac{\omega_{CaCO_3,inventory}}{M_{CaCO3}}\right)}; \qquad [s^{-1}]$$

The solids circulation rate describes the transition of a potentially active mol of CaO on its way between carbonation and calcination within the overall gasifier system. The **bed material renewal rate** is a further important key figure for the overall process performance. Following equation is used for the calculation of the bed material renewal rate:

$$(2) \qquad \kappa_{CaO} = \frac{\dot{m}_{bed,fresh} \cdot \left(\frac{\omega_{CaO,bed,fresh}}{M_{CaO}} + \frac{\omega_{CaCO_3,bed,fresh}}{M_{CaCO3}}\right)}{m_{inventory} \cdot \left(\frac{\omega_{CaO,inventory}}{M_{CaO}} + \frac{\omega_{CaCO_3,inventory}}{M_{CaCO3}}\right)}; \qquad [s^{-1}]$$

Additional fresh bed material is necessary to maintain the **CO_2 transport capacity** of CaO particles because of the decay of transport capacity after N cycles as shown in **Fig. 1**. Therefore, fresh $CaCO_3$ is fed to the bed material circulation loop just before the combustion and calcination step. A high bed material renewal rate raises the CO_2 transport capacity of the bulk bed material in circulation, increases the selective transport of CO_2 from the gasifier but at the same time reduces the cold gas efficiency. The CO_2 transport capacity of the bed material is therefore a further central key figure for the process. The following equations are used for the calculation of the CO_2 transport capacity based on a mass and molar perspective:

(3) $$\varphi_{CaO} = \frac{\dot{m}_{CO2,capt}}{\dot{m}_{bed,G,in} \cdot \left(\omega_{CaO,bed,G,in} + \omega_{CaCO_3,bed,G,in} \cdot \frac{M_{CaO}}{M_{CaCO3}} \right)}; \qquad \left[\frac{kg_{CO2}}{kg_{CaO}}\right]$$

(4) $$X_{CaO} = \frac{\frac{\dot{m}_{CO2,capt}}{M_{CO2}}}{\dot{m}_{bed,G,in} \cdot \left(\frac{\omega_{CaO,bed,G,in}}{M_{CaO}} + \frac{\omega_{CaCO3,bed,G,in}}{M_{CaCO3}} \right)}; \qquad \left[\frac{mol_{CO2}}{mol_{CaO}}\right]$$

High CO_2 transport capacity improves the selective transport of CO_2 from the gasifier to the combustion chamber and therefore, increases the product gas quality by reducing the CO_2 content of the product gas. The described equations have been implemented in IPSEpro and enable a discussion of relevant parameters within the process simulation environment based on existing experimental data.

Fig. 2 shows the approach of the used simulation model. Within the created simulation model relevant mass streams are modeled as represented by arrows in **Fig. 2**. Variations should show the energetic behavior of biomass steam gasification by sorption enhanced reforming. The variation steps are carried out with respect to achievable cold gas efficiency, product gas composition, bed material consumption and key parameters for the design of experimental facilities.

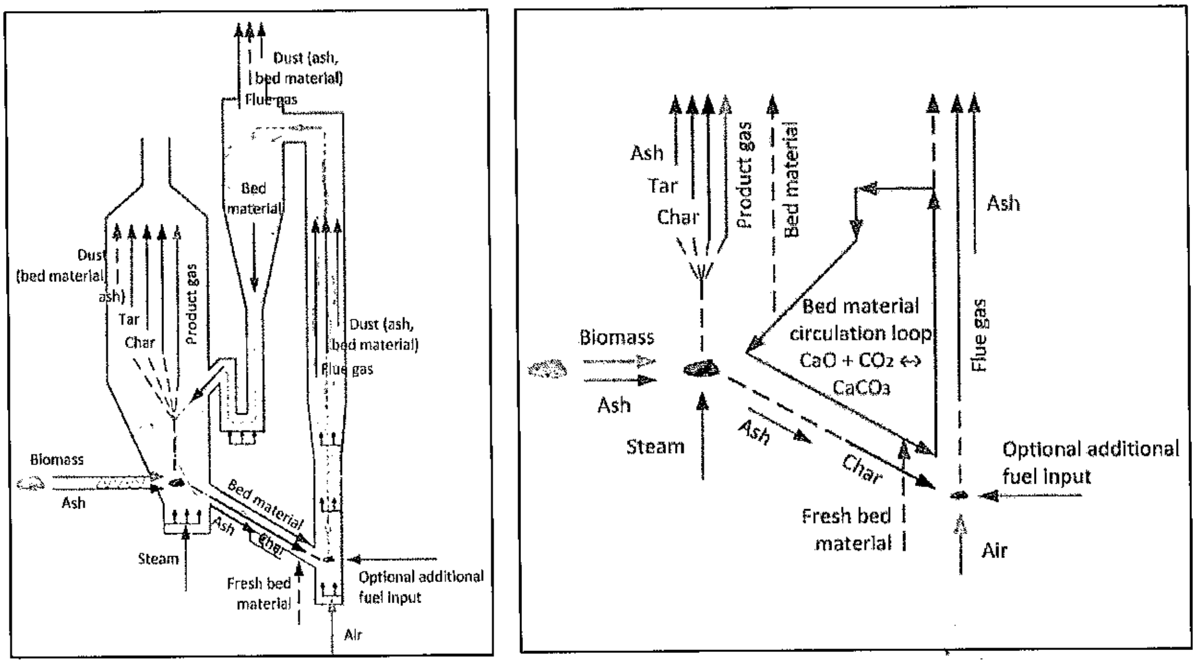

Fig. 2: Schematic representation of mass streams calculated within the IPSEpro simulation model of gasification by sorption enhanced reforming. Illustration based on Schmid, J.C. (2012)

The following important equations have been used additionally within the simulation model:

(5) $$10^{\delta_{Eq.WGS-shift}} \cdot K_p(T) = \frac{p_{CO2} \cdot p_{H2}}{p_{CO} \cdot p_{H2O}} \; [-];$$

(6) $$\tau_{CaO} = \frac{1}{\kappa_{CaO}} \; [s];$$

(7) $$N = \tau_{CaO} \cdot \delta_{CaO} \; [-];$$

(8) $$\eta_{chem} = \frac{P_{pg}}{P_{th}} \; [-];$$

Table 2: Variations carried out within the created simulation model

Nr.	Variation parameters	Sym.	Unit	Values	Results	Additional constants[1]
1	CO_2 transport capacity	X_{CaO}	$[mol_{CO2}/mol_{CaO}]$	0.0 - 0.55	Fig. 4 (b), Fig. 5 (b), Fig. 6 (a), Fig. 6 (b)	κ_{CaO} = 0.065* *equals $\dot{m}_{bed,fresh}$ = 200 kg/h
2	CO_2 transport capacity bed material renewal rate	X_{CaO} κ_{CaO}	$[mol_{CO2}/mol_{CaO}]$ $[h^{-1}]$	0.1 - 0.8 0.0 - 1.0	Fig. 4 (a)	-
3	CO_2 transport capacity mean cycle number	X_{CaO} N	$[mol_{CO2}/mol_{CaO}]$ [-]	0.0 - 0.85 5 – 100	Fig. 5 (a)	-

1 Variations are based on reference case in Table 1. Additional constants are added for a single variation to enable a specific investigation case

RESULTS AND DISCUSSION

Fig. 3 shows the flow chart of the created simulation model based on the simplified modeling approach shown in **Fig. 2**. Several variations illustrated in Table **2** based on the reference case shown in **Table 1** were carried out.

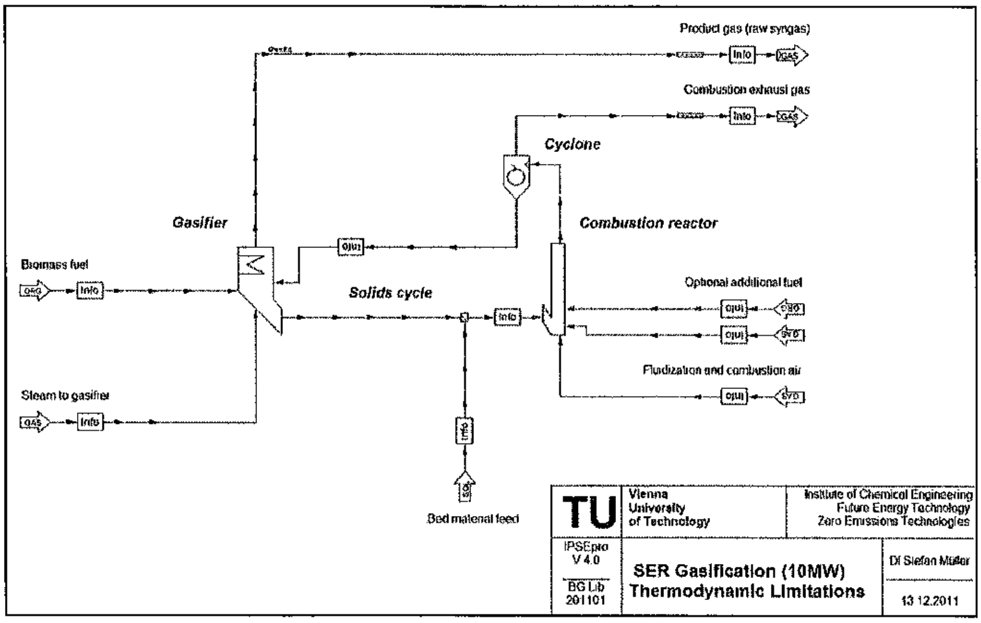

Fig. 3: Simulation model of sorption enhanced reforming in dual fluidized bed gasification system

Cold Gas Efficiency & Product Gas Composition

Achievable cold gas efficiency is strongly determined by temperatures, fuel composition, fuel water content and heat losses. Cold gas efficiency is furthermore depending on the bed material renewal rate as shown in **Fig. 4** (a). Large amounts of fresh $CaCO_3$ entering the system raise the heat demand in the combustion chamber for calcination and maintaining the necessary temperatures. Furthermore, low CO_2 transport capacity of the circulating bed material raises the necessary amount of circulating bed material to maintain the process. According to the described relationships bed material renewal rates should be kept on a low level with in terms of cold gas efficiency. The composition of the product gas is strongly depending on the selective transport of CO_2 from the gasifier to the combustion chamber. Therefore, the CO_2 transport capacity of the circulating bed material has a major influence on the product gas composition. This dependency is shown in **Fig. 4** (b). The equilibrium of the water-gas-shift reaction has

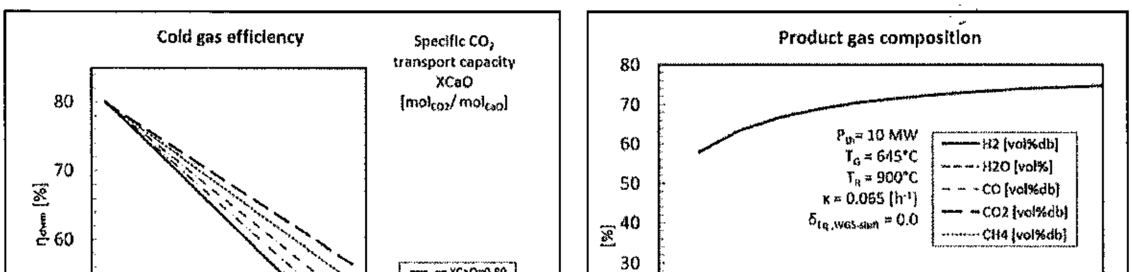

Fig. 4: (a - left) Cold gas efficiency depending (η_{chem}) on bed material renewal rate, (b - right) Product gas composition depending on CO_2 transport capacity of circulating bed material.

been an important precondition for the calculation of the composition of the product gas at the exit of the gasifier. Solid particles with high CO_2 transport capacity would raise the H_2 content in the product gas. At the same time high bed material renewal rates would be necessary to keep the transport capacity at high levels with respect to the findings of Grasa et al. (2006) shown in **Fig. 1**. Favorable product gas compositions and high cold gas efficiency can therefore be identified as an important trade-off of gasification with sorption enhanced reforming. An improvement of product gas composition by the operation of higher bed material renewal rates leads to a reduction of cold gas efficiency. In specific cases this reduction of the cold gas efficiency can be prevented by arranging the process along industrial processes involving a certain lime and/or limestone throughput.

Bed Material Consumption

CO_2 transport capacity of circulating solid particles is a limiting parameter for the overall process. The practical performance of different lime stones has a strong influence on **bed material consumption**. Low transport capacity after low cycle numbers (N) leads to a high demand for bed material renewal. The practical performance of solid particles directly influences bed material consumption and cold gas efficiency. **Fig. 5** (a) shows necessary bed material renewal rates for different transport capacities of lime stone particles after different mean cycle numbers completed. A real system includes bed material inventory which consist of different fractions of solid particles which experienced a different number of cycles. Within the simulation model the occurring cycle number distribution of the operating bed material is represented by the parameter N (mean cycle number). Furthermore, it has been assumed that solid particles are ejected at random and old solid particles are not discharged selectively from the operating bed material. This simplified approach has to be acknowledged at the evaluation of the results.

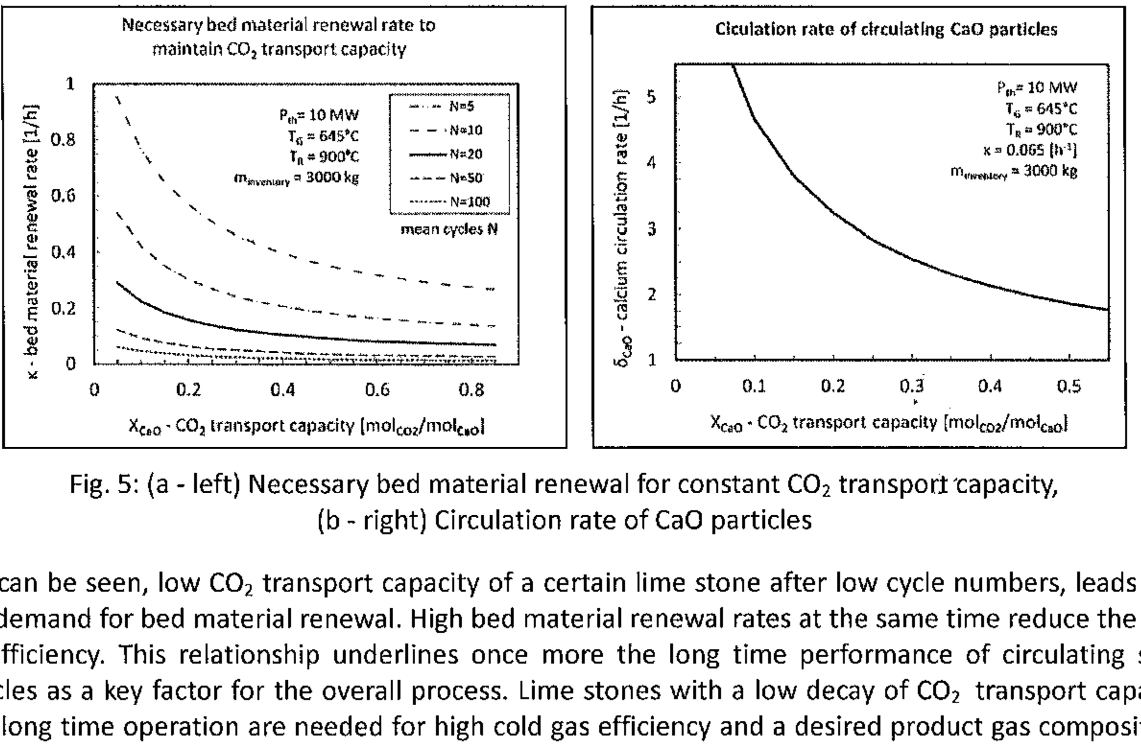

Fig. 5: (a - left) Necessary bed material renewal for constant CO_2 transport capacity,
(b - right) Circulation rate of CaO particles

As it can be seen, low CO_2 transport capacity of a certain lime stone after low cycle numbers, leads to a high demand for bed material renewal. High bed material renewal rates at the same time reduce the cold gas efficiency. This relationship underlines once more the long time performance of circulating solid particles as a key factor for the overall process. Lime stones with a low decay of CO_2 transport capacity after long time operation are needed for high cold gas efficiency and a desired product gas composition. Otherwise high bed material consumption could be a significant disadvantage for the realization of sorption enhanced reforming. **Fig. 5** (b) additionally shows the calcium circulation rate for solid CaO particles with different transport capacity. High CO_2 transport capacities enable a lower cycling rate because necessary heat for the gasification is transported in form of latent heat together with bound reaction energy of the carbonation reaction. Simulation results shown in **Fig. 5** (b)show necessary circulation rates which were calculated for a fixed bed material renewal rate of $\kappa = 0.065$ per hour. This value equals 200 kg of fresh bed material per hour according to the set reference case. A low bed material renewal rate is desired to keep cold gas efficiency high and bed material consumption low. Attrition and or decay of CO_2 transport capacity can be limiting parameters which demand for higher bed material renewal rates. This tradeoff therefore has a strong influence at the necessary circulation rate depend on solids particle performance. The attrition of solid particles is at the same time strongly dependent on the used solids separation systems, the mechanical stability of used bed material and velocities maintained within the dual fluidized bed system.

Key Parameters for the Design of Experimental Facilities

Relevant key parameters for the design of facilities operating the investigated process are **volume flows** and **bed material circulation** in kg/h. Volume flows need to be considered during the dimensioning of the dual fluidized bed system hosting the process. **Fig. 6** (a) shows the influence of the selective transport of CO_2 from the gasification reactor to the combustion reactor.

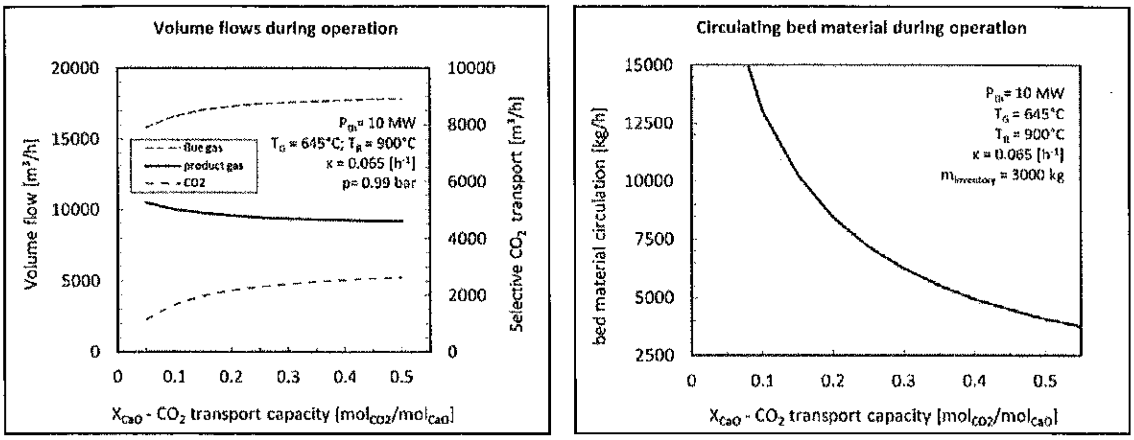

Fig. 6: (a) Volume flows during operation, (b) Amount of circulating bed material during operation

As it can be seen, selective transport of CO_2 has only a small impact on the operational volume flows. Nevertheless, changes of volume flows need to be considered regarding the influence on the fluid dynamics within the system. **Fig. 6** (b) shows the necessary bed material circulation for the gasification by sorption enhanced reforming for different CO_2 transport capacities of the operating bed material. As can be seen from **Fig. 6** (b), the bed material circulation is strongly influenced by CO_2 transport capacity. This relationship should be considered before the design of experimental facilities.

CONCLUSIONS

The achieved results highlight main relationships within the process of gasification by sorption enhanced reforming. A simple model reveals important aspects of the process. The decay of CO_2 transport capacity with an increased number of carbonation/calcinations cycles has been previously identified as a key factor for the performance of gasification by sorption enhanced reforming. The optimum bed material circulation rate is determined by the energy balance of the system requiring the temperature levels for carbonation and calcination. As a consequence, the transport capacity of the sorbent directly determines the achievable product gas composition. In order to compensate the decay of CO_2 transport capacity after several calcination and carbonation cycles, continuous addition of fresh bed material is necessary to maintain a certain desired product gas composition. Significant bed material make-up rates, however, reduce in most cases the cold gas efficiency. It is important to note that the use of lime/limestone in biomass gasification shows advantages such as low tar content even if the selective CO_2 transport is moderate. A certain make-up rate will always be required in order to compensate for attrition. Low mechanical stability of the used bed material therefore can be a further constraint for the process which would lead to reduced cold gas efficiency. Mechanical stability of different lime stones itself (Koppatz et al. 2009) has been beyond the focus of present work. But, the effect of necessary bed material renewal on cold gas efficiency due to attrition can be estimated with the results shown in **Fig. 4**. Further research should identify realistic process scenarios considering both, capacity decay and attrition. Additional, reduced cold gas efficiency can be prevented in specific cases. If high H_2 contents are the primary aim, the process could be arranged along with industrial processes involving a certain limestone throughput.

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ABBREVATIONS

A_G	[m²]	Cross-sectional area of gasifier
$K_p(T)$	[-]	Equilibrium constant
$□_{bed,fresh}$	[kg/h]	Fresh bed material ($CaCO_3$) entering bed material circulation loop
$□_{bed,G,in}$	[kg/h]	Massflow of bed material entering the gasifier from combustor
M_{CaCO3}	[g/mol]	Molar weight of $CaCO_3$
M_{CaO}	[g/mol]	Molar weight of CaO
M_{CO2}	[g/mol]	Molar weight of CO_2
$□_{CO2,capt}$	[kg/h]	Massflow of CO_2 absorbed by bed material within gasifier
$□_{fluid,G}$	[kg/h]	Fluidization mass flow gasifier (steam)
$m_{gasifier}$	[kg]	Amount of bed material within gasifier
$m_{inventory}$	[kg]	Total bed material within system
N	[-]	Mean cycles of particle within system
P_A	[kW]	Optional additional fuel power
p_{CO}	[Pa]	Partial pressure of CO in product gas
p_{CO2}	[Pa]	Partial pressure of CO_2 in product gas
p_{H2}	[Pa]	Partial pressure of H_2 in product gas at
p_{H2O}	[Pa]	Partial pressure of H_2O in product gas
P_{PG}	[kW]	Product gas power, LHV based, (Boie)
P_{th}	[kW]	Thermal fuel power, LHV based
Q_{loss}	[kW]	Heat loss of overall system
$T_{fluid,G}$	[°C]	Fluidization steam temperature
$T_{fluid,R}$	[°C]	Fluidization air temperature
T_G	[°C]	Gasification temperature
T_R	[°C]	Combustion temperature
X_{CaO}	[mol_{CO2}/mol_{CaO}]	CO_2 transport capacity
δ_{CaO}	[s^{-1}]	Calcium circulation rate
$\delta_{Eq.,WGS\text{-}shift}$	[-]	Logarithmic distance to equilibrium (water-gas-shift reaction, gasifier)
η_{chem}	[-]	Cold gas efficiency
κ_{CaO}	[s^{-1}]	Bed material renewal rate
λ_R	[-]	Air ratio for combustion
σ_{CO}	[mol/mol]	CO / CO_2 in combustor exhaust gas
τ_{CaO}	[s]	Mean time of particle within system
$□_{CaO}$	[kg_{CO2}/kg_{CaO}]	CO_2 transport capacity
$\omega_{CaCo3,bed,fresh}$,	[$kg_{CaCO3}/kg_{bed,fresh}$],	$CaCO_3$ in fresh material
$\omega_{CaCo3,bed,G,in}$,	[kg_{CaCO3}/kg_{bed}],	$CaCO_3$ in bed material to gasifier from combustor
$\omega_{CaCo3,inventory}$,	[kg_{CaCO3}/kg_{bed}],	$CaCO_3$ in bed mat. inventory
$\omega_{CaO,bed,fresh}$,	[$kg_{CaO}/kg_{bed,fresh}$],	CaO in fresh bed material
$\omega_{CaO,bed,G,in}$,	[kg_{CaO}/kg_{bed}],	CaO in bed material to gasifier from combustor
$\omega_{CaO,inventory}$,	[kg_{CaO}/kg_{bed}],	CaO in bed mat. inventory

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