Evaluation of Sorption Enhanced Reforming in Combination with Oxyfuel Combustion for the Sequestration of CO₂

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Abstract (short summery / version of your already submitted abstract)

Traditional dual fluid gasification enables a favorable product gas comprising hydrogen, carbon monoxide, carbon dioxide, and methane. This paper focus on Sorption Enhanced Reforming combined with Oxyfuel combustion, which constitutes an additional opportunity for CO₂ sequestration. First experimental results in the test plants at TU Wien (100 kW) and at University Stuttgart (200 kW) have been implemented. Based on these results, the functionality of OxySER could be demonstrated and a concept for the commercial-scale use is developed. Furthermore, the profitability of OxySER, by application in a raw iron production plant for the generation of reduction gas, within a techno-economic assessment is investigated. The results of the techno-economic assessment show that the production of reduction gas via Sorption Enhanced Reforming in combination with Oxyfuel combustion can definitely compete with the natural gas route. The biggest potential to increase the profitability of the OxySER process offers the use of low-grade fuels like bark or sugarcane bagasse to decrease the fuel costs, which is the main cost driver.

1. Introduction:

Since industrialized nations consume more energy per capita than developing countries as well as the increasing standard of living in general raise the future challenges for developing a sustainable economic system based on renewable technologies. Additionally, future challenges of climate change and the exorbitant carbon dioxide (CO₂) emissions from fossil fuels lead to a high demand of CO₂-neutral technologies in future. Since biomass releases the same amount of CO₂ as it aggregates during its growth, the utilization of biogenic fuels can contribute significantly to a reduction of CO₂ emissions. Further, several technologies provide the possibility of additional carbon-emission reduction by sequestration of CO_2 from industrial processes.

Fig. 1 represents the way towards CO₂ sequestration within an industrial plant in the context of a combustion process and further CO₂ utilization or storage. The biggest potential for the sequestration of CO₂ are in the raw iron production, refinery, cement production and the natural gas processing. [1] Within these plants, a sequestration process can be implemented. Post combustion capture, Pre combustion capture, Oxyfuel combustion and Separated combustion are several possibilities for industrial CO₂ sequestration. Further explanations regarding CO₂ sequestration can be found in [1], [2] and [3].



Fig. 1: Possibilities for CO₂ sequestration [2]

The main focus of the paper lies on Oxyfuel combustion. This CO_2 sequestration technology uses as fluidization agent a mix of pure oxygen and recirculated flue gas. Therefore, the nitrogen from the air is excluded from the combustion system. [2]

Furthermore, the sequestrated and purified CO₂ could be used in further process steps as raw material, **Carbon capture and utilization (CCU)**, or stored in underground deposits, **Carbon capture and storage (CCS)**. [3], [4]

Today around 130 million tons of carbon dioxide are utilized materially. 110 million tons are used as product and 20 million tons as industry gas. [4]

Beside the CCU technologies, CO_2 can also be stored in underground deposits. CCS is banned in Austria except research projects up to a storage volume of 100 000 t of CO_2 . [6]

1.1 Sorption Enhanced Reforming

A promising option for the sequestration of CO_2 from biomass and the generation of a hydrogen-rich product gas at the same time is the Sorption Enhanced Reforming process in combination with Oxyfuel combustion (**OxySER**).

The Sorption Enhanced Reforming (SER) is based on the dual fluidized bed steam gasification process. Detailed information regarding the dual fluidized bed steam gasification process can be found in literature. [5]–[7] **Fig. 2** demonstrates the basic principle of SER.



Fig. 2: Concept of Sorption Enhanced Reforming [8]

First of all, biomass, residues or waste materials are introduced in the gasification reactor. Limestone is used as bed material which serves as transport medium for heat, but also as carrier for CO₂ from the gasification (GR) reactor to the combustion reactor (CR) by adjusting the temperature levels in the reactors correctly. Within the SER process, steam serves as fluidization and gasification

agent in the GR. Therein, several endothermic gasification reactions take place in a temperature range between 600-700°C. [5] Residual char is transferred with the bed material from the GR to the CR. The CR is operated within a temperature range between 800-900°C and with the fluidization agent air. By combustion of residual char in the CR, heat is released. This suitable temperature profiles in the GR and CR ensure that the bed material (limestone) is first calcined to calcium oxide (CaO) at high temperatures in the CR (2). Then the CaO is carbonized in the GR with the carbon dioxide from the product gas (1). Thus, in this cyclic process a transport of CO₂ from the product gas to the flue gas appears. [9] The use of steam in the gasification reactor and the water gas shift reaction (3) in combination with in-situ CO₂ sorption via the bed material system CaO/CaCO₃ enables the production of a nitrogen free and hydrogen enriched product gas as showed in **Tab. 1**. [5], [10]

Carbonation (exo.): $Ca0 + CO_2 \rightarrow CaCO_3$ (1) Calcination (endo.): $CaCO_3 \rightarrow Ca0 + CO_2$ (2) WGS (exo.): $CO + H_2O \rightarrow CO_2 + H_2$ (3)

Parameter	Unit	Conventional gasification (100 kW)	Gasification by SER (100 kW)
Fuel		wood pellets	wood pellets
Bed material		olivine	limestone
Particle size	mm	0.4 - 0.6	0.5 - 1.3
Water (H ₂ O)	vol%	30 - 45	50 - 65
Hydrogen (H ₂)	vol% _{dry}	36 - 42	55 - 75
Carbon monoxide (CO)	vol%dry	19 - 24	4 - 11
Carbon dioxide (CO ₂)	vol% _{dry}	20 - 25	6 - 20
Methane (CH ₄)	vol% _{dry}	9 - 12	8 - 14
Non cond. hydrocarbons (C _x H _y)	vol% _{dry}	2.3 - 3.2	1.5 - 3.8
Dust particles	g/Nm ³	10 - 20	20 - 50
Tar	g/Nm ³	4 - 8	0.3 - 0.9

Tab. 1: Comparison product gas composition of conventional gasification and SER [5]

The gasification decrease of the temperature comparison to in the gasification, leads conventional to increasing demands on the bed material cooling. [11]

Tab. 1 represents a comparison between product gas compositions the of conventional gasification and SER. As mentioned above, the carbon dioxide content of the product gas could be reduced through the SER method. Furthermore, the hydrogen content is higher in comparison to the conventional gasification. The catalytic activity of limestone enables a reduction of tar at the same time. [4], [5], [12]

1.2 Oxyfuel combustion

The Oxyfuel combustion is characterized by the use of a mixture of pure oxygen and recirculated flue gas as oxidation agent. Fig. 3 represents the concept of Oxyfuel combustion. The reason for the flue gas recirculation is the related possibility of temperature regulation in the combustion chamber. In case of application of Oxyfuel combustion on dual fluidized bed steam gasification, a mixture $(\dot{m}_{0_2,fluid})$ of 30 vol.-%_{dry} pure oxygen ($\dot{m}_{O_2,clean}$) and 70 vol.-%_{dry} flue gas $(m_{O_2,recirc})$ has been determined based on a variety of tests on the pilot plant at the TU Wien. [2] This is a good compromise between safety, efficiency and technical requirements. [2]



Fig. 3: Concept of Oxyfuel combustion [2]

After condensation of steam, a CO_2 enriched flue gas stream is produced. An important operating parameter is the oxygen surplus ($\dot{m}_{O_2,Combustion}$). The aim of this technology is to reach low oxygen contents and high CO₂ contents in the flue recirculation The of flue gas. gas $(m_{O_2,recirc})$ implies also the recirculation of surplus oxygen. Therefore, the stoichiometric combustion ratio of the combustion step ($\lambda_{Combustion}$) is always higher than the stoichiometric combustion ratio of the overall process ($\lambda_{process}$). [2]

1.3 Combination of Oxyfuel combustion and Sorption Enhanced Reforming

The combination of Oxyfuel combustion and Sorption Enhanced Reforming (OxySER) combines the advantages of both technologies. **Fig. 4** represents the concept of the combined technology. [4] Compared with **Fig. 2**, which shows the SER process, the combustion part is now operated as Oxyfuel combustion as explained in Chapter 1.2.



Fig. 4: Concept of OxySER [8]

By the use of renewable fuels and a continuous sequestration and storage or utilization of CO_2 , an improved CO_2 balance can be achieved. [4], [13] **Tab. 2** represents test results of the 200 kW pilot plant at University Stuttgart. Therein, product gas compositions from the SER and OxySER gasification technology are compared. The comparison illustrates that a CO_2 enriched flue gas in the OxySER test rig in Stuttgart was obtained.

Parameter	Parameter Unit		Gasification by OxySER (200 kW)
Fuel		wood pellets	wood pellets
Bed material		limestone	limestone
Particle size	mm	0.3 - 0.7	0.3 - 0.7
Р	roduct gas cor	nposition	
Water (H ₂ O)	vol%	50	50
Hydrogen (H ₂)	vol% _{dry}	69 - 72	70
Carbon monoxide (CO)	vol% _{dry}	8 - 11	8
Carbon dioxide (CO ₂)	vol% _{dry}	5 - 7	8
Methane (CH ₄)	vol% _{dry}	11 - 12	11
Non cond. hydrocarbons (C _x H _y)	vol% _{dry}	2 - 3	3
Tar	g/Nm ³	14	6
	Flue gas comp	position	
Water (H ₂ O)	vol%	14	30
Oxygen (O ₂)	vol% _{dry}	7	9
Nitrogen (N ₂)	vol% _{dry}	46	-
Carbon dioxide (CO ₂)	vol% _{dry}	47	91

Tab. 2: Comparison product gas and flue gas composition of SER and OxySER [13]

However, OxySER implies the following advantages in comparison to the conventional gasification:

- selective CO₂ transport to flue gas,
- decrease of tar content in product gas,
- high CO₂ content in flue gas > 90 vol.-%_{dry}[13],
- smaller flue gas stream because of flue gas recirculation,
- nitrogen free flue gas.

These assumptions serves as a basis for the conception of an industrial application.

2. Concept and methodology:

With regard to the technoeconomic assessment of the CO_2 sequestration technology OxySER, a plant concept for the integration in an existing industrial plant has been developed.

2.1 Concept of CO₂ sequestration with OxySER

The OxySER plant concept for integration in an existing raw iron production plant is illustrated in **Fig. 5**. The plant shows a product gas power of 10 MW. For the production of 10 MW product gas, 5000 kg/h of wood chips with a water content of 40 wt.-% are required. [5] The wood chips are treated in a biomass dryer. Afterwards the biomass is fed in the gasification reactor. The bed material inventory (limestone) of the system contains 2500 kg. In the gasification reactor, a H₂ enriched product gas with a temperature of 680 °C is produced. Subsequently, the dust particles are removed out of the product gas by a cyclone. Besides ash, these dust particles contain still carbon. This is the reason why the particles are recirculated to the combustion reactor. [4]



Fig. 5: OxySER plant concept with 10 MW product gas power for the integration in an existing industry plant [5]

Afterwards, the product gas is cooled down to 180°C. The released heat can be used for preheating of the biomass dryer air. [4] Furthermore, the product gas filter seperates further fine dust particles out of the product gas stream and conveys them back to the combustion reactor. In the gas cleaning system of the existing raw iron plant, tar is seperated in a scrubber and water is condensed. Biodiesel is used as solvent. The product gas exits the scrubber with a temperature of 40°C. Afterwards, it compressed in а blower and is subsequently used as reduction gas in a blast furnance. The CO₂ enriched flue gas leaves the combustion reactor with a

temperature of 900°C. The flue gas is cooled down to 180°C by the steam superheater and a flue gas cooler. Steam is heated up to 450°C in a countercurrent heat exchanger. Fly ash is removed out of the system by a flue gas filter. A partial flow from the flue gas is recirculated and mixed with pure oxygen. Pure oxygen is produced by an air seperation unit. The remaining flue gas stream is compressed in the flue gas blower and water is condensed in a flue gas dryer. The cleaned CO₂ rich gas can be used in different CCU processes. [4] The integration approach offers the advantage to use existing equipment, like gas cleaning steps and air separation units from industrial plants. Furthermore, the produced product gas can be used directly in the industrial plant, for example as reduction gas in raw iron production plants. [4]

2.2 Simulation of mass- & energy balances with IPSEpro

Mass and energy balances of the concept of CO_2 sequestration in combination with OxySER, based on model libraries, which were developed on the TU Wien, has been calculated in IPSEpro. For further information regarding IPSEpro a reference is made to [14] and [15].

2.3. Techno-economic assessment with net present value calculation

techno-economic The assessment regarding the net present value (NPV) calculation serves as decision-making tool for the valuation of upcoming investments. The NPV is a function of the investment and operating costs. The operating costs are multiplied by the cumulative present value factor, which includes the interest rate and the plant lifetime. Therefore, the calculation helps to compare NPV expected payments in the future with current payments. Further information can be found in [16] and [7].

3. Results and discussion

Based on experiences of the pilot plant from the TU Wien and the University of Stuttgart, combined with the previously described concept, the mass- and energy balances were calculated. Furthermore, the mass- and energy balances are the basis for a techno-economic assessment. **Tab. 3** and **Tab. 4** represents the plant utilities and operating parameters of an OxySER plant.

Parameter	Unit	Value	Ref.
Bed material inventory	kg	2500	[5]
Fuel (wood chips)	kg/h	5040	[5]
Fresh bed material	kg/h	177	[16]
Cooling capacity in % of fuel power	% (kW/kW _{th})	5 - 20	[11]
Oxygen	Nm ³ /h	1102	[5]
Fresh water	kg/h	37.8	[5]
RME	kg/h	20	[5]
Flushing gas	Nm ³ /h	50	[5]

Tab. 3: Plant utilities of an OxySER plant
with 10 MW product gas energy

Tab. 5 shows the exiting mass- and volume flows of an OxySER plant with 10 MW product gas energy. It can be seen, that 5040 kg/h of wood chips and $1102 \text{ Nm}^{3/\text{h}}$ of pure oxvgen are required for the production of 2880 Nm³/h product gas. The product gas is used as reduction gas in the raw iron production route. Furthermore, 3610 kg/h of CO₂ can be produced for further utilization. The costs for final disposal of 105 kg/h of ash and dust have been taken into account.

Parameter	Unit	Value	Ref.
Lower heating value (wood chips)	MJ/kg	9.53	[5]
Water content (wood chips)	wt%	40	[5]
Combustion temperature	°C	900-950	[17]
Gasification temperature	°C	625-680	[17]
Particle size (bed material)	μm	375-550	Assumption
Coarse ash	μm	375-550	Assumption
Fine ash	μm	< 100	Assumption
Very fine ash	μm	< 20	Assumption
Water content (product gas)	vol%	7.81	[5]
Lower heating value (product gas)	MJ/kg	22.88	[5]

Tab. 4: Operating parameters of an OxySER plant with 10 MW product gas energy

Parameter	Unit	Value	Ref.
Product gas	Nm ³ /h	2880	IPSE
Flue gas	Nm ³ /h	5300	IPSE
Ash and dust	kg/h	105	[5]
Bed material	kg/h	100	[4]
Carbon dioxide (for CCU)	kg/h	3610	[5]

Tab. 5: Output flow streams of an OxySER plant with 10 MW product gas energy

The techno-economic assessment relies on the results of the IPSEpro simulation. **Tab. 6** represents the fuel prices for chosen fuel types. It is thus evident that natural gas is with $25 \notin$ /MWh (see **Tab. 6**) the most expensive fuel within this analysis and sugarcane bagasse with $6 \notin$ /MWh the cheapest fuel.

Fuel type	Unit	Value	Ref.
Wood chips	€/MWh	22.5	[4]
Natural gas	€/MWh	25	[18]
Bark	€/MWh	19.2	[16]
Sugarcane bagasse	€/MWh	6	Assumption

Tab. 6: Fuel prices for techno-economic assessment

In **Tab. 7** and **Tab. 8** cost rates for utilities and for the NPV calculation are listed. Exemplary, the costs for one employee per year are assumed to $57\ 000\ \text{€/a}$ and the expected plant lifetime of an OxySER plant is 20 years. (see **Tab. 8**)

Parameter	Unit	Value	Ref.
Electricity	€/kWh	0.04	[16]
Limestone	€/t	35	[16]
Nitrogen	€/Nm ³	0.003	[16]
Fresh water	€/t	0.02	[16]
Solvent (RME)	€/t	960	[16]
Oxygen (air separator)	€/Nm ³	0.02	Assumption
Oxygen (external purchase)	€/Nm ³	0.07	[5]
Emission allowances certificate	€/t _{CO2}	23	[19]
Costs for ash disposal	€/t	90	CHP Güssing
CO ₂ expenses	€/Nm ³	0.06	Assumption

Tab. 7: Cost rates for utilities

Parameter	Unit	Value	Ref.
Maintenance costs per year	%/a	2.00	[7]
Insurance, administration & tax per year	%/a	2.50	[7]
Number of employees (Integration)	-	3	Assumption
Costs of one employee per year	€/a	57000	[16]
Expected plant life time	а	20	Assumption
Interest rate	%	4	Assumption

Tab. 8: Cost rates for NPV calculation

The techno-economic analysis is based on the following **business case** that an operator of a raw iron production plant would like to build a new reduction gas supply unit driven by biogenic feedstock. The NPV calculation, which is shown in **Tab. 9**, serves as decision-making tool. The goal to produce 10 MW reduction gas, should be achieved with regard to CO_2 emissions. The reference option (Option 0) is the production of reduction gas by steam reforming of natural gas. Furthermore, various alternative options (Options 1-4) are compared with the reference option:

- **Option 0** (reference case): Production of 10 MW reduction gas through steam reforming of natural gas,
- **Option 1:** Production of 10 MW reduction gas through gasification of wood chips by SER,
- **Option 2**: Production of 10 MW reduction gas through gasification of wood chips by OxySER,
- **Option 3**: Production of 10 MW reduction gas through gasification of bark by OxySER,
- **Option 4**: Production of 10 MW reduction gas through gasification of sugarcane bagasse by OxySER.

The SER process in Option 1 requires no pure oxygen for operation. However, the flue gas of the SER process cannot be exploited in further utilization steps because of the high nitrogen content in the flue gas. The alternative Options 2-4 are based on the SER process in combination with Oxyfuel combustion. The shared use of gas cleaning and utility preparation steps already existing in steel industry, like air separation, decrease the investment costs by 50% (assumption). The OxySER process is based on the assumption that the CO_2 is sold as product for utilization. **Tab. 9** represents the Net present value calculation for the production of 10 MW reduction gas. Therein, the fuel energy per year, the investment costs including interest and fuel costs per year are listed. Furthermore, **Tab. 9** shows beside the fuel costs also all other consumption related costs. Costs for CO_2 emission certificates are paid only for the use of fossil fuels (reference case).

Parameter	Unit	Natural gas	SER wood chips (Integration)	OxySER wood chips (Integration)	OxySER bark (Integration)	OxySER bagasse (Integration)
		Reference	Option 1	Option 2	Option 3	Option 4
Product gas energy	MW	10	10	10	10	10
Fuel energy natural gas	MWh/a	70 000				
Fuel energy wood chips	MWh/a		93 100	93 100		
Fuel energy bark	MWh/a				93 100	
Fuel energy sugarcane bagasse	MWh/a					93 100
Investment costs incl. interest	€	8 000 000	10 000 000	10 000 000	10 000 000	10 000 000
Fuel costs natural gas	€/a	1.750.000				
Fuel costs wood chips	€/a		2 094 750	2 094 750		
Fuel costs bark	€/a				1 787 520	
Fuel costs sugarcane bagasse	€/a					558 600
CO ₂ emission certificates	€/a	322 000				
Maintenance, insurance, etc.	€/a	360 000	450 000	450 000	450 000	450 000
Employee costs	€/a	57 000	171 000	171 000	171 000	171 000
Auxiliaries	€/a	33 215	177 770	177 770	177 770	177 770
Disposal costs	€/a		66 150	66 150	66 150	66 150
Oxygen costs	€/a			154 280	154 280	154 280
Sum of Expenses per year	€/a	2 522 215	2 959 670	3 113 950	2 806 720	1 577 800
Earnings CO ₂	€/a			765 758	765 758	765 758
Sum of Earnings per year	€/a			765 758	765 758	765 758
Investment costs incl. interest	€	8 000 000	10 000 000	10 000 000	10 000 000	10 000 000
Expenses - Earnings	€/a	2 522 215	2 959 670	2 348 193	2 040 963	812 043
Additional investment costs (compared to reference option)	€	0	2 000 000	2 000 000	2 000 000	2 000 000
Operating expenses savings	€/a	0	- 437 455	174 022	481 252	1 710 172
Relative Net Present Value	€	0	- 7 945 017	364 963	4 540 219	21 241 241





Fig. 7: Relative Net Present Value



Fig. 6: Main costs per year

The Relative NPV represents the profitability of alternative production routes in comparison to the reference route. The NPV of Option 1 shows a negative value. This means that the operation of SER with wood chips based on the expected plant lifetime of 20 years is less profitable than the reference option. techno-economic comparison The between SER and OxySER shows that the earnings through carbon dioxide are much higher than the oxygen costs. This is the reason for the positive NPV and thereby profitable operation in Option 2. Due to less fuel costs in the alternative Options 3 and 4, the profitability increase. Fig. 7 represents the main costs of all options per year. It can be seen, that the fuel costs are the main cost driver in the process.

Additionally, a sensitivity analysis of fuel prices, cost rates for utilities and cost rates for NPV have been created. **Tab. 10** represents the sensitivity analysis based on Option 3 of the techno-economic assessment. The cost rates are calculated for the case that the NPV for Option 3 is equal to the reference option.

Parameter	Unit	Initial Value	Sensitivity Value NPV = 0	Dev.	
Bark	€/MWh	19.2	22.8	19%	
Natural gas	€/MWh	25.0	20.2	-19%	
Investment costs	€	10.0 Mio	12.8 Mio	28%	
Oxygen (air separator)	€/Nm ³	0.02	0.06	200%	
Solvent (RME)	€/t	960	3350	249%	
Costs for ash disposal	€/t	90	544	504%	
Limestone	€/t	35	304	769%	
	Fuel costs (very sensitive)				
	Investment costs (sensitive)				
	Utility costs (not sensitive)				

Tab. 10: Sensitivity analysis cost rates

The sensitivity values, shown in **Tab. 10**, are the limit values for the profitability of Option 3 compared with the reference option. This means for example that Option 3 is favourable compared to the reference option if the investment costs does not exceed 12.8 Mio \in .

4. Conclusion and Outlook

The gasification via SER allows the in-situ CO_2 sorption via the bed material system $CaO/CaCO_3$. Therefore, the selective transport of carbon dioxide to the flue gas stream is reached. The use of a mix of pure oxygen and recirculated flue gas as fluidization agent in the CR results in a nearly pure CO_2 flue gas stream. The CO_2 could be used for further synthesis processes like e.g. the urea synthesis.

The results of the techno-economic assessment show that the production of reduction gas via Sorption Enhanced Reforming in combination with Oxyfuel combustion can definitely compete with the natural gas route. Especially, lowgrade fuels like bark or sugarcane bagasse increase the profitability of the OxySER process. Furthermore, the sensitivity analysis of the cost rates show that the fuel and investment costs have more influence on the profitability of the OxySER plant than the utility costs. This means, to increase the profitability of a DFB plant, further research work should focus on the development of additional integration process routes with the use of low-grade fuels.

Summing up, the shown integration concept indicates valuable data for the design of the proposed concept. Beforehand a demonstration at a reasonable scale is recommended.

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LIST OF ABBREVIATIONS

LIST OF SYMBOLS

CCS	Carbon Capture and Storage	$\lambda_{Combustion}$	stoichiometric combustion
CCU	Carbon Capture and Utilization	Combustion	ratio of the combustion step
CaCO ₃	calcium carbonate	2	stoichiometric combustion
CaO	calcium oxide	reprocess	ratio of the overall process
CH ₄	methane	m .	mass flow of pure oxygen
CO	carbon monoxide	m0 ₂ ,clean	mass flow of pure oxygen
CO_2	carbon dioxide	m _{O2} ,combustion	mass now of oxygen in the
CR	combustion reactor		flue gas
C _x H _y	non condensable hydrocarbons	$m_{O_2,fluid}$	mass flow of oxygen in the
Dev.	deviation		oxidation agent
dry	dry basis	$\dot{m}_{O_2,recirc}$	mass flow of oxygen in
GŘ	gasification reactor		recirculated flue gas
H ₂	hydrogen	0 _{2,rest}	remaining oxygen in the
H ₂ O	water		flue gas after recirculation
NPV	Net Present Value	ω	recirculation rate
OxySER	Sorption Enhanced Reforming in		
•	combination with Oxyfuel		
	combustion		
Ref.	reference		
SER	Sorption Enhanced Reforming		

vol.-% volumetric percent

wt.-% weight percent

WGS water gas shift reaction

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