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Dedicated to Professor Liviu Literat, at his 80th anniversary

GASIFICATION PROCESS – A PRACTICAL WAY FOR SOLID FOSSIL FUELS DECARBONISATION

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ABSTRACT. This paper investigates the technical aspects of solid fossil fuels decarbonisation by gasification process. More specifically, it focuses on the technical evaluation and the assessment of performance of an Integrated Gasification Combined Cycle (IGCC) scheme modified for Carbon Capture and Storage (CCS). One of the key units of such a plant is the gasification island, a number various commercial gasification reactors (mainly entrained-flow types of dry feed and slurry feed) are evaluated for decarbonisation of fossil fuels (coal).

The significant issues in the overall design of fossil fuels decarbonisation process using gasification process are: proper choice of gasification reactor and feedstock (e.g. fluxing or fuel blending), downstream syngas treatment options (shift conversion of carbon monoxide, syngas desulphurization and later carbon dioxide capture) and finally the usage of combustible decarbonised gas for various energy vectors poly-generation (e.g. power, H_2 etc.).

Finally, whole process for one case study (using Siemens gasification reactor) was mathematical modeled and simulated using ChemCAD software for the particular case of hydrogen and electricity co-production scheme based on coal with carbon capture and storage.

Keywords: *Fossil fuels decarbonisation, Gasification, Carbon Capture and Storage*

INTRODUCTION

Gasification of solid fuels (either only fossil fuels or in addition with various renewable energy sources such biomass or solid wastes) forms the core of Integrated Gasification Combined Cycle (IGCC) concepts which is one of the advanced clean coal power generation systems [1, 2]. Compared with conventional coal fired power plants, IGCC scheme has lower emissions of acid gases (NO_X, SO_X) and particulate matter. In addition, with respect to clean

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coal technology, IGCC has a better carbon dioxide capture potential, using pre-combustion capture whereas conventional coal fired power plants have to use post-combustion capture [2, 3].

The IGCC is an energy conversion process concept in which the solid fuel (e.g. coal) is gasified to produce a fuel gas (called syngas), which after purification (ash removal and desulphurization), is burnt in the gas turbine in a conventional Combined Cycle Gas Turbine (CCGT) to produce power. Because the gasification process produces a large amount of waste heat, which can be used for steam production, the steam systems in the CCGT heat recovery steam generator and gasification system are integrated together, hence the IGCC appellation (the combination of two thermodynamic cycles namely Brayton cycle and Rankine cycle) [4].

In gasification processes designed for fuel decarbonisation, oxygen plus steam or water is reacted with coal at high temperatures to produce syngas which mainly consists of carbon monoxide and hydrogen. After purification performed to remove ash and sulphur compounds, the mixture of CO and H_2 is normally used as a fuel gas for the gas turbine (no capture option). But for the capture option (pre-combustion capture of carbon dioxide), the hydrogen level is raised by catalytic conversion of carbon monoxide in the syngas with steam, the chemical reaction being:

$$CO \quad + \quad H_2O \quad \leftrightarrow \quad H_2 \quad + \quad CO_2$$

This step also enables the carbon present in the coal to be concentrate in form of carbon dioxide that can be later captured. After shift conversion, the gas, which now contains mainly carbon dioxide and hydrogen, is cooled to ambient temperature then sent to an Acid Gas Removal (AGR) system for CO_2 and H_2S removal. Here, CO_2 and H_2S are removed by chemical solvents similar with those used in post-combustion capture (alkanolamines such as methyl-diethanol-amine - MDEA) as well as by physical solvents (e.g. Rectisol®, Selexol®) [5].

After AGR, the hydrogen-rich gas stream can be used for various purposes: to produce hydrogen for chemical and petro-chemical processes (e.g. ammonia and methanol synthesis, hydrogenation, hydrocracking and hydrodesulphurization processes etc.) or for co-production of hydrogen and electricity. In this last option, one of the main advantages of such a scheme is the possibility to run the plant full-load all the time. It is well known that power duty has daily, weekly and seasonal variations and the power generation has to follow the duty (demand). In such a case (e.g. during the night), the power plants have to run part-load or even to be switched off which implies severe reduction of the plant lifetime (thermal stress, corrosion etc.) and negative impact on plant economic indicators.

The co-production mode of hydrogen and electricity has the merits that during the low power duty periods, the plant can produce hydrogen which, apart from power, can be stored. In this case the plant is running full load all time with significant benefits in term of plant lifetime and economic indicators. In co-production mode, the hydrogen-rich gas is split in two: one stream is purified by Pressure Swing Adsorption (PSA) and then is sent to the hydrogen customers and rest is used in a CCGT for power generation.

This paper investigates the technical aspects of coal gasifier selection for co-production of electricity and hydrogen based on IGCC processes with carbon capture and storage [6-9]. More specifically, it focuses on the technical evaluation and the assessment of performance of a number of coal gasifiers based on existing moving-bed, fluidised-bed and entrained-flow gasification processes.

The key factors considered in the analysis are: gasifier throughputs, reliability and experience, gasifier pressure and temperature, cold gas efficiency (CGE), carbon conversion, water or steam requirement, downstream gas clean up issues, hydrogen production potential, implication of oxygen purity for hydrogen purification stage, implication of gasifier selection for AGR system etc. Based on these key factors, a list of the most promising coal gasifiers for electricity and hydrogen co-production based on IGCC processes with carbon capture is proposed.

After analyzing the most promising gasification reactors for coal, for one case the whole IGCC concept with carbon capture and storage will be modeled and simulated using ChemCAD software for generation of about 400 MW power and a flexible hydrogen output between 0 and 200 MW hydrogen (considering the hydrogen lower heating value – LHV) with about 90 % carbon capture rate (decarbonisation rate).

Plant concept and gasifier options for hydrogen and electricity co-production with carbon capture

An IGCC plant modified for co-production of hydrogen and electricity with carbon capture and storage is basically similar to that of existing IGCC plants for producing electricity only without carbon capture. The main differences are the need for shift conversion stage to increase hydrogen production and to concentrate the carbon species in form of carbon dioxide, an Acid Gas Removal (AGR) system which in addition to hydrogen sulphide (as in conventional technology) is capturing also carbon dioxide and a hydrogen purification stage by PSA.

A simplified flowsheet of the hydrogen and electricity co-production (HYPOGEN concept) plant based on coal with carbon dioxide capture is presented in the Figure 1 [10]. In this case, based on an entrained-flow design, a syngas boiler is used to recover waste heat before the syngas is sent to the shift converter. Other entrained-flow gasifiers dispense with this option and quench the gas with water to solidify and remove slag.

As shown, carbon dioxide stream is compressed to pressures more than 100 bar before being sent to storage site (geological storage or for Enhanced Oil Recovery - EOR). This also gives a difference between the type of plant and a conventional IGCC, the compression of carbon dioxide requiring a significant amount of energy which implies an energy penalty.

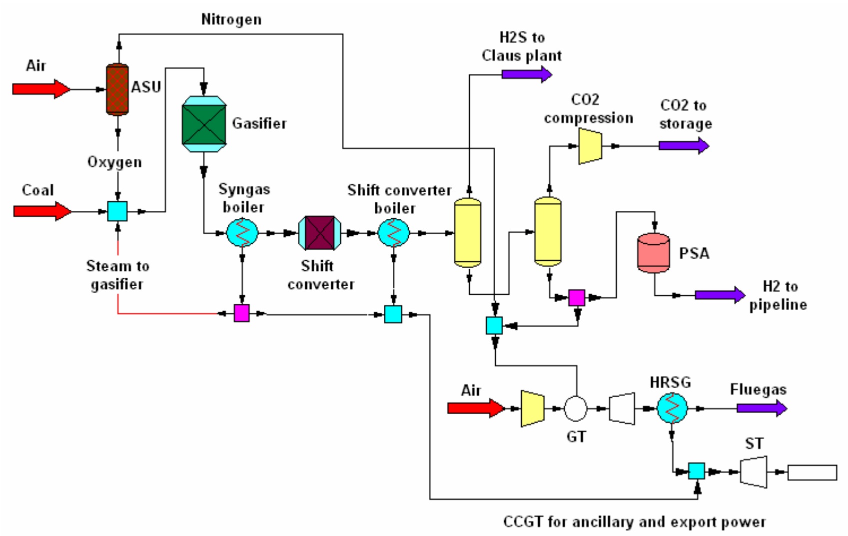

Figure 1. Hydrogen and electricity co-production scheme with carbon capture and storage based on coal gasification.

An other main difference is the addition of a Pressure Swing Absorption (PSA) system for purifying the hydrogen outlet stream of the plant. Because the need to find suitable solutions for replacing fuel oil-derived products (gasoline and diesel) for transport sector, the hydrogen stream purity is set to 99.95 % (vol.) to be used in Proton Exchange Membrane (PEM) fuel cells designed for mobile applications.

Gasification reactors can be grouped into the following three main categories: moving-bed gasifiers (sometimes also called fixed-bed gasifiers), fluidised-bed gasifiers and entrained-flow gasifiers [1, 10-11]. A short description of each of these categories is discussed below:

- *Moving-bed gasifiers* are characterized by a bed in which the fuel moves slowly downward under gravity as it is gasified. The phases circulation is a counter-current one. In such a counter-current arrangement, the hot gases from the gasification zone are used to preheat and pyrolyse the downward flowing coal. With this type of gasifier, the oxygen consumption is low but pyrolysis products are present in the syngas and also the outlet temperatures are low (400 – 600^{o}C). Within this category there are two different gasifier types: the dry ash type in which the temperature is kept below the ash melting point, and the slagging type in which the temperature in the gasifier heart is above the ash melting point.

The main shortcoming of this type of gasifier is that a considerable amount of methane will be produced, which reduces the amount of hydrogen that can be generated and decrease the carbon capture capabilities of the plant (methane cannot be captured by AGR) [10].

- *Fluidized-bed gasifiers* in which the bed is maintained in a fluidised condition by an upward flow of gaseous stream. This configuration offers extremely good mixing between feed and the oxidant, which promotes both heat and mass transfer. This ensures an even distribution of material in the bed, and hence a certain amount of partially un-reacted coal is removed with the ash (lower carbon conversion compared with other gasifier types). This gasifier type has moderate oxygen and steam consumptions [1].

Unfortunately operation of fluidised beds are restricted to temperatures below ash softening point (900 – 1050^{o}C), as operation above this temperature would cause the ash to agglomerate. This implies the need for a highly reactive fuel. Coal is therefore not suitable as much fuel would remain in an un-reacted condition. Methane also is a problem for fluidised beds and this feature, plus the loss of carbon in the ash, results in this process being less ideal for hydrogen production or carbon capture.

- *Entrained-flow gasifiers* are reactors in which the particles of coal are relatively dispersed and are carried along by a blast of reactants and products in co-current flow. The residence time is just a few seconds and the temperatures are in the range of 1250 – 1600^{o}C to ensure good carbon conversion. High temperature ensures that the syngas has very low methane content and because the temperature is high and the fuel is dry, there are no limitations on the type of coal.

The method of introducing the fuel into the gasifiers is specific to the design. There are, basically, two ways in which coal is transported into an entrained-flow gasifier: via water slurry (slurry feed gasifier), and via a gas, typically nitrogen (dry feed gasifier). For the slurry feed gasifier, the need to bring the water in the slurry up to gasifier temperatures, results in some of the coal having to be combusted, producing CO_2, as this reaction provides much more energy than the reaction of coal with oxygen to produce CO. The effect of this is to reduce the amount of hydrogen that can be produced, since the CO can subsequently react with steam to form hydrogen. As a conclusion regarding the gasification reactor feed system, the dry feed system ensures a higher energy efficiency of the gasifier [1].

The key factors that are considered for selection of gasifiers for hydrogen and electricity co-production with CO_2 capture are [1,10]: oxygen purity (usually 95 % O_2 vol., air blown gasifiers being unsuitable because of the nitrogen dilution), gasifier throughputs, reliability and experience on the industrial scale (400 MW power), cold gas efficiency (CGE) and carbon conversion (which are optimum in case of entrained-flow gasifiers), syngas cooling options (the quench type being desirable because the steam requirement of the shift conversion), hydrogen production potential (hydrogen and carbon monoxide content as high as possible in the syngas) and implication of gasifier reactor on Acid Gas Removal system (from this point of view a gasifier able to run at high pressure being desirable).

Considering the various commercial gasification reactors and their characteristics [1, 10-11] and analyzing these reactors against the above criteria, the most promising options for hydrogen and electricity co-generation schemes with carbon capture and storage are:

- Entrained-flow gasifier with dry feed and heat recovery (developed by Shell);
- Entrained-flow gasifier with dry feed and water quench (developed by Siemens formerly known Future Energy GmbH);
- Entrained-flow gasifier with slurry feed and water quench (developed by General Electric – Texaco).

The next section of this paper presents in detail a detailed modeling study for a plant concept based on Siemens gasification technology (entrained-flow gasifier with dry feed and syngas quench). The choice of Siemens gasification technology for an IGCC scheme with carbon capture for hydrogen and power generation is based on the following main factors: dry feed design which implies an increase energy efficiency compared with slurry feed design, water quench of the hot syngas which ensure the optimum condition for the shift conversion (no extra steam has to be added to the syngas), good cold gas efficiency and carbon conversion, good hydrogen production potential and clean syngas.

RESULTS AND DISCUSSIONS

This section presents a modeling study of a whole IGCC plant designed for hydrogen and electricity co-production with carbon capture based on Siemens gasification reactor. For the case study presented in detail below, the coal characteristics considered as feedstock are presented in Table 1.

The main sub-systems of the plant for co-generation of hydrogen and power co-generation with carbon capture and theirs design assumptions used in the mathematical modeling and simulation are presented in Table 2 [2, 8,12-15].

Table 1. Feedstock (coal) characteristics

Parameter	Coal
Proximate analysis (% wt)	
Moisture	8.10
Volatile matter	28.51
Ash	14.19
Ultimate analysis (% wt dry)	
Carbon	72.04
Hydrogen	4.08
Nitrogen	1.67
Oxygen	7.36
Sulphur	0.65
Chlorine	0.01
Ash	14.19
Lower heating value - LHV (MJ/kg a.r.)	25.353

Table 2. Main design assumptions for the plant concept

Unit	Parameters
Air separation unit (ASU)	Oxygen purity: 95 % (vol.) Delivery pressure: 2.37 bar Power consumption: 225 kWh/ton O_2 No integration with gas turbine
Gasifier (Siemens)	Pressure: 40 bar Pressure drop: 1.5 bar Temperature: >1400^oC Water quench
Shift conversion (WGS)	Sulphur tolerant catalyst Two adiabatic beds Pressure drop: 1 bar / bed
Acid gas removal (AGR)	Solvent: Selexol® (mixture of di-methyl ethers of poly-ethylene glycols) Separate capture of CO_2 and H_2S Solvent regeneration: - thermal (heat) for H_2S step - pressure flash for CO_2 step
CO_2 compression and drying	Delivery pressure: 100 bar Compressor efficiency: 85 % Solvent used for drying: TEG (Tri-ethylene-glycol) Drying level: < 500 ppm H_2O
Claus plant & tail gas treatment	Oxygen-blown H_2S-rich gas composition: > 20 % (vol.) Tail gas recycled to H_2S absorption stage
Pressure Swing Adsorption (PSA)	Purified hydrogen: > 99.95 % (vol.) Purification yield: 85 % Tail gas pressure: 1.5 bar (recycled to the power island)
Gas turbine	Gas turbine type: M701G2 (Mitsubishi Heavy Industries Ltd.) Net power output: 334 MW Electrical efficiency: 39.5 % Pressure ratio: 21 Turbine outlet temperature (TOT): 588^oC
Heat recovery steam generation (HRSG) and steam cycle	Three pressure levels: 118 bar / 34 bar / 3 bar Reheat of MP steam Steam turbine isoentropic efficiency: 85 % Steam wetness ex. steam turbine: max. 10 %
Heat exchangers	$\Delta T_{min.}$ = 10^oC Pressure drop: 1 % of inlet pressure

The hydrogen and electricity co-generation plant based on Siemens gasification technology presented in Figure 1 was modeled and simulated using ChemCAD software. The case study was simulated in different situations (only electricity or various modes of hydrogen and electricity co-production). In case of power generation only mode, the gas turbine is running full load and for hydrogen and electricity co-production mode the gas turbine is gradually turned down to 80 % in order to displace an energy stream in form of hydrogen-rich gas which can be then purified in a Pressure Swing Adsorption (PSA) unit to produce a purified hydrogen stream (99.95 % vol.) to be used in transport sector (PEM fuel cells).

Table 3 presents the overall plant performance indicators (gross and net power output, electrical and hydrogen efficiencies, specific CO_2 emissions) in electricity only and hydrogen and electricity co-generation with carbon capture and storage.

Table 3. Overall plant performance indicators

Main Plant Data	Units	Power	Power + hydrogen		
Coal flowrate (a.r.)	t/h	165704			
Coal LHV (a.r.)	MJ/kg	25.353			
Feedstock thermal energy – LHV (A)	MW_{th}	1166.98			
Syngas thermal energy (B)	MW_{th}	934.75			
Cold gas efficiency (B/A * 100)	%	80.10			
Thermal energy of syngas ex. AGR (C)	MW_{th}	830.70			
Syngas treatment efficiency (C/B *100)	%	88.86			
Gas turbine output (1 x M701G2)	MW_e	334.00	314.97	296.27	277.58
Steam turbine output (1 ST)	MW_e	197.50	187.44	177.38	167.40
Expander power output	MW_e	0.78	0.72	0.66	0.61
Gross electric power output (D)	MW_e	532.28	503.13	474.31	445.59
Hydrogen output – LHV (E)	MW_{th}	0.00	50.00	100.00	150.00
ASU consumption + O_2 compression	MW_e	44.72	44.72	44.72	44.72
Gasification island power consumption	MW_e	8.08	8.08	8.08	8.08
AGR + CO_2 drying & compression	MW_e	40.07	40.07	40.07	40.07
H_2 compression	MW_e	0.00	0.66	1.33	2.01
Power island power consumption	MW_e	19.00	18.30	17.55	16.80
Total ancillary power consumption (F)	MW_e	111.87	111.83	111.75	111.68
Net electric power output (G = D - F)	MW_e	420.41	391.30	362.56	333.91
Gross electrical efficiency (D/A * 100)	%	45.61	43.11	40.64	38.18
Net electrical efficiency (G/A * 100)	%	36.02	33.53	31.06	28.61
Hydrogen efficiency (E/A * 100)	%	0.00	4.28	8.57	12.85
Cumulative efficiency (G+E/A * 100)	%	36.02	37.81	39.63	41.46
Carbon capture rate	%	92.35	92.35	92.35	92.35
CO_2 specific emissions	kg/MWh	76.12	80.16	86.27	93.41

Compared with classic IGCC technology for power generation without carbon capture which has an overall net energy efficiency of about 42 % [16], the modified IGCC technology for carbon capture is penalized by 7 – 8 % which is the energy penalty for the carbon capture process. This is the price in term of energy for decreasing the specific CO_2 emissions from about 800 kg/MWh (for no capture case) to about 76.12 kg/MWh for the capture case (92.35 % carbon capture rate).

Another fact that has to be mentioned for hydrogen and power co-production mode, is the overall energy efficiency of the plant that is increasing in the situation in which the ancillary power consumption is remaining virtually constant (see Table 3). This fact is very important and attractive for plant cycling (modification of the power generated by the plant according to the demand of the national grid) considering that for low electricity demand the plant can produce mostly hydrogen which compared with power can be stored to be used either for covering the peak loads or for other applications (transport sector, petro-chemical sector etc.).

Figure 2 presents the variations of net electrical efficiency, hydrogen efficiency and overall energy efficiency with hydrogen and power co-production rate (thermal energy of the hydrogen output stream).

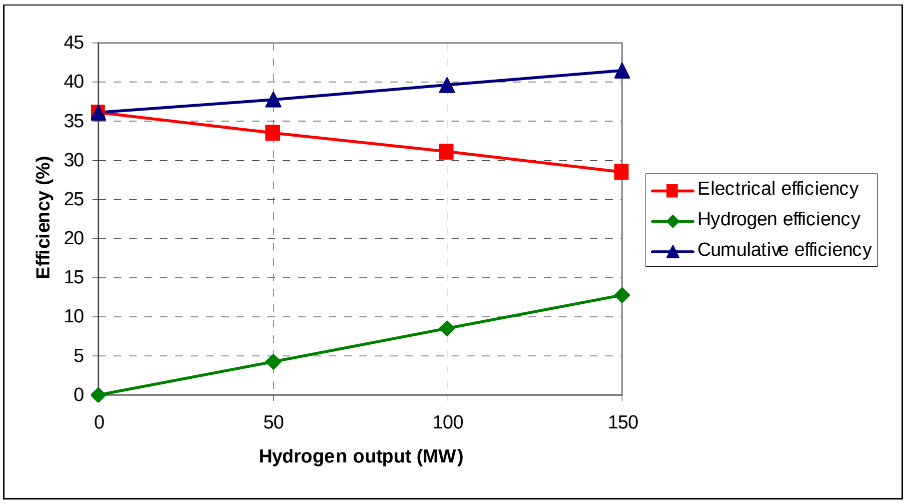

Figure 2. Variation of net electricity, hydrogen and overall efficiency vs. hydrogen and power co-production rate.

In the range of 0 to 200 MW hydrogen (based on 10.795 MJ/Nm^3 hydrogen lower heating value), the gas turbine can be gradually turned down to about 80 % of the nominal load [4]. For higher hydrogen plant flexibility (for the case described above, i.e. more than 200 MW hydrogen), another approach must be used in plant design namely building a separate power block designed to provide the ancillary power for the plant [17, 18].

CONCLUSIONS

This paper assesses the main characteristics of coal gasifiers for hydrogen and electricity co-production scheme with carbon capture based on a modified IGCC plant design. The main aim has been to develop evaluation criteria and then to select the most appropriate coal gasification concepts for hydrogen and electricity co-production with carbon capture and storage (CCS) and to investigate how the gasifier selection affects the other sub-systems of the plant (e.g. Acid Gas Removal unit).

The most three promising concepts for hydrogen and electricity co-production with carbon capture based on coal gasification are all based on entrained-flow gasifiers (dry feed with heat recovery - Shell, dry feed with water quench – Siemens and slurry feed with water quench – GE Texaco).

The paper also presents in detail a case study for hydrogen and electricity co-production based on Siemens gasifier and with carbon capture step using a physical absorption (solvent Selexol®). This case study was modeled and simulated using ChemCAD software and the simulation results were used for quantification of main performance characteristics (overall plant efficiency, carbon capture rate, specific CO_2 emissions etc.).

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