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ASSESSMENT OF FLEXIBLE CARBON CAPTURE AND UTILIZATION OPTIONS APPLIED TO GASIFICATION PLANTS

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ABSTRACT. The aim of this work is to assess the energy vector poly-generation capabilities of gasification plants equipped with carbon capture and utilization (CCU) features. As evaluated energy carriers, various total or partial decarbonized vectors were investigated (e.g., power, hydrogen, synthetic natural gas, methanol, Fischer-Tropsch fuel). As illustrative examples, the gasification concepts with 100 MW net energy output were considered having an overall plant decarbonization rate of 90%. As decarbonization technologies, the gas – liquid absorption based on chemical and physical scrubbing was assessed. A broad range of process system engineering tools were used (e.g., modeling and simulation, process integration, plant flexibility elements, technical and environmental evaluation). As results show, the application of carbon capture and utilization technologies for gasification-based poly-generation has promising results in term of increasing the overall energy efficiency (up to 68%), reducing CO_2 emissions (down to 7 kg/MWh) and improving cycling capabilities.

Keywords: *Carbon capture and utilization (CCU) technologies, Gasification, Energy vectors poly-generation, Technical and environmental assessment.*

INTRODUCTION

Greenhouse gas emissions (especially CO_2) represent a significant issue of the modern world. Global warming and climate change are caused by increased anthropogenic greenhouse gas emissions compared to pre-industrial levels [1]. Important technical, economic, social and political efforts

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are devoted to tackle these significant environmental issues. In this respect, the energy-intensive industrial applications (e.g., heat and power generation, chemical, metallurgical and cement sectors) should be significantly re-design to curb greenhouse gas emissions for future low-carbon economy as well as to improve the overall energy efficiency [2].

Several technical methods are already available for developing the future low-carbon economy e.g., increasing the share of renewable energy sources (e.g., wind, solar, biomass), improving the energy conversion and utilization aspects, developing Carbon Capture, Utilization and Storage (CCUS) applications [3-4]. Since for the heat and power generation, suitable renewable solutions are already in place (e.g., wind mills, thermal and photovoltaic solar systems), for other important energy-intensive and polluting sectors such as chemical, petro-chemical, iron and steel production, cement production, the suitable solutions are still to be developed considering the particular characteristics of these systems. For non-power applications, the renewable energy sources (e.g., wind and solar) have a limited applicability, the conventional fuels (either fossil or renewable) being a more suitable solution [5].

CCUS technologies have a promising development potential since they can be successfully used to make environmental acceptable even the most polluting fossil fuels (e.g., coal, lignite, oil). Carbon capture and utilization methods are aiming to mitigate the carbon dioxide emissions from various industrial applications and then to utilize the captured CO_2 in different ways: production of synthetic fuels, mineralization for construction materials, raw material for organic synthesis [6-7].

Along these important lines, the present paper is aiming to evaluate the potential energy vectors poly-generation based on gasification process. Various total and partial decarbonized energy carriers (e.g., power, hydrogen, substitute natural gas, synthetic liquid fuels) were assessed to be produced based on syngas processing. The following syngas-based reactions are used for energy vectors poly-generation [8-10]:

- Hydrogen production via water gas shift (WGS) conversion:

$$CO + H_2O \rightarrow CO_2 + H_2 \tag{1}$$

- Synthetic natural gas (SNG) production via methanation:

$$CO + 3H_2 \rightarrow CH_4 + H_2O \tag{2}$$

- Methanol production:

$$CO \ + \ 2H_2 \ \rightarrow \ CH_3OH \tag{3}$$

- Fischer-Tropsch synthesis:

$$nCO \ + \ \left(\frac{m}{2}+n\right)H_2 \ \rightarrow \ C_nH_m \ + \ nH_2O \tag{4}$$

To reduce the carbon dioxide emissions, a carbon capture technology (based on chemical and physical scrubbing) was also fitted in the gasification plant. The overall concepts are characterized by improved overall energy efficiency and low CO_2 emissions. As an illustrative example, chemical gas-liquid abruption using Methyl-Di-Ethanol-Amine (MDEA) was considered according to the following reaction [11]:

$$CO_2 \ + \ MDEA \ + \ H_2O \ \rightarrow \ MDEAH^+ \ + \ HCO_3^- \tag{5}$$

In addition to decarbonization, the syngas-based poly-generation concept has important advantages in improving the plant cycling capabilities. In energy sector, the current fossil-based facilities are under increasing pressure to be re-design to make them more flexible in order to accommodate the time-irregular renewable energy sources. In this respect, a flexible poly-generation concept, which can produce electricity during peak times and other energy carriers (various chemicals) during periods with low electricity demand, is of great importance [12-13].

LAYOUT OF GASIFICATION-BASED POLY-GENERATION CONCEPT AND MAIN DESIGN ASSUMPTIONS

The conceptual design of flexible and decarbonized coal-based gasification plant is presented in Figure 1 [14]. Coal is gasified with oxygen and steam leading to syngas which is furthermore cooled down and the ash is removed. Subsequently, a water gas shift conversion is necessary to increase the hydrogen content simultaneously with reduction of carbon monoxide content to the molar ratio required for various reactions. Carbon dioxide and hydrogen sulfide are captured separately in an Acid Gas Removal (AGR) unit followed by flexible poly-generation step of total or partial decarbonized energy carriers (electricity, hydrogen, synthetic fuels).

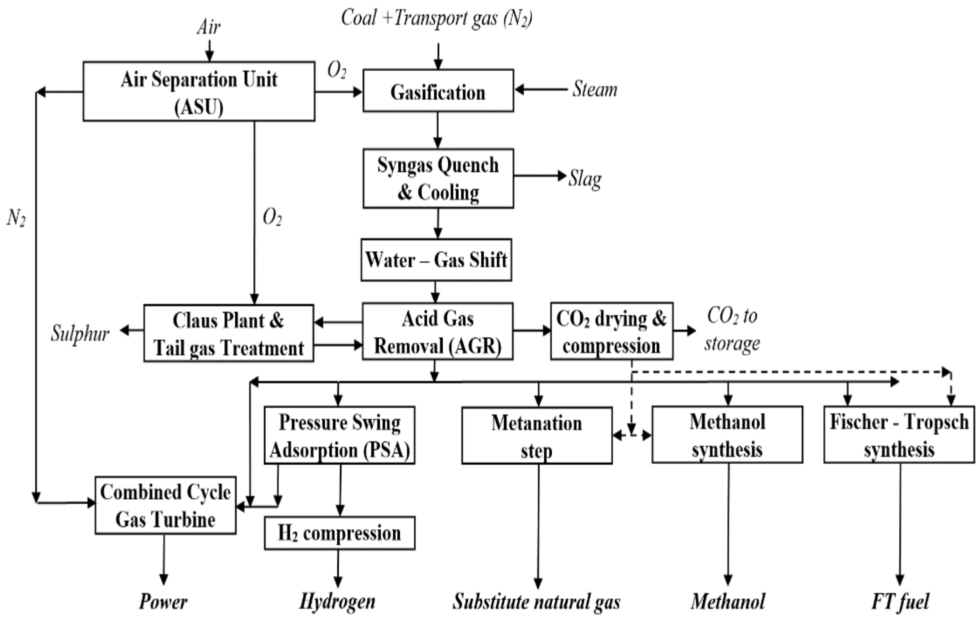

Figure 1. Flexible and decarbonized gasification-based energy vector poly-generation concept

The proposed concept was evaluated in a flexible energy vector poly-generation scenario to generate 100 MW net energy output (for the main energy carrier) with a 90% carbon capture rate for AGR unit. The gasification-based system was modeled and simulated using ChemCAD software. Table 1 presents the main design assumptions used in the evaluation [9,14].

For syngas-processing units into various energy carriers (SNG, methanol, FT fuel), the conventional configurations and process operation conditions based on literature sources [15-18] were considered.

Table 1. Main design assumptions of flexible poly-generation concept

Plant sub-system	Design assumptions
Coal characteristics	Ultimate analysis (% wt. dry): 72.04% carbon, 4.08% hydrogen, 1,67% nitrogen, 7.36% oxygen, 0.65% sulphur, 0.01% chloride, 14.19% ash; Moisture: 8% Lower heating value (LHV): 25.35 MJ/kg
Air separation unit	Purity (% vol.): 95% O_2, 3% N_2, 2% Ar Power consumption: 200 kWh/t oxygen Oxygen delivery pressure: 2 bar
Gasification unit	Shell reactor (dry fed gas quench) Operating pressure: 40 bar Operating temperature: 1400 – 1500°C Pressure drop: 1 bar Gas quench temperature: 800°C
Water Gas Shift (WGS) unit	No. of catalytic beds: 2 Reactor type: equilibrium Thermal mode: adiabatic Steam to CO ratio: 2 (molar) Pressure drop: 1 bar / catalytic bed
Chemical CO_2 capture unit Absorption column: Desorption column:	Solvent: Methyl-DiEthanol-Amine (MDEA) Solution concentration: 50 % wt. No. of stages: 20 Column pressure drop: 1 bar No. of stages: 10 Column pressure drop: 1 bar Solvent regeneration: thermal (LP steam) Heat duty: 0.65 MJ/kg CO_2
Physical CO_2 capture unit Absorption column:	Solvent: Selexol^{TM} (mixture of methyl ethers of poly-ethylene glycol) No. of stages: 20 Column pressure drop: 1 bar Solvent regeneration: pressure reduction (4 stages)
Claus plant	Type: oxygen-fed Inlet gas composition (vol.): > 25% H_2S Sulphur recovery: > 99%
CO_2 processing unit	Drying agent: Tri-Ethylene-Glycol (TEG) 4 compressing stages with inter-cooling Delivery pressure: 120 bar CO_2 composition (vol. %): >95% CO_2, <2000 ppm CO, <250 ppm H_2O, <100 ppm H_2S, <4% other gases (N_2, Ar, H_2)
Power block	Combined cycle gas turbine Net electrical efficiency: 39.5% Pressure ratio: 21 HP / MP / LP steam levels: 120 / 34 / 3 bar Steam turbine efficiency: 85% Condensing pressure: 48 mbar Cooling water temperature: 15°C
Hydrogen processing unit	Delivery pressure: 60 bar Compressor efficiency: 85% Outlet temperature: 30-40°C
Heat exchangers	$\Delta T_{min.}$ = 10°C; Pressure drop: 2 - 3% of inlet pressure

EVALUATION METHODOLOGY

Various energy vectors poly-generation systems were modeled and simulated using ChemCAD software. In selection of the thermodynamic package, the chemical compounds as well as the operating conditions were considered. For instance, in case of gas processing units (e.g., syngas conditioning, physical gas-liquid absorption, chemical reactors for synthesis of various energy carriers), Soave-Redlich-Kwong (SRK) package was used. For chemical gas-liquid absorption, the electrolyte package was used based on the present ionic system. In case of captured CO_2 conditioning system (CO_2 drying), TEG Dehydration package was selected. For steam generation and power block, Thermoflex software was chosen to double-check the simulation results obtained in ChemCAD.

The assessed process configurations were optimized in term of energy utilization by thermal integration analysis. In this aim, the pinch analysis was used to evaluate the overall hot and cold utility consumptions [19]. As an illustrative example, Figure 2 presents the hot and cold composite curves for gasification-based system used for decarbonized power generation (combined cycle power block). The results derived from simulation were compared with available literature and experimental data for model validation [20-21]. No significant differences were observed.

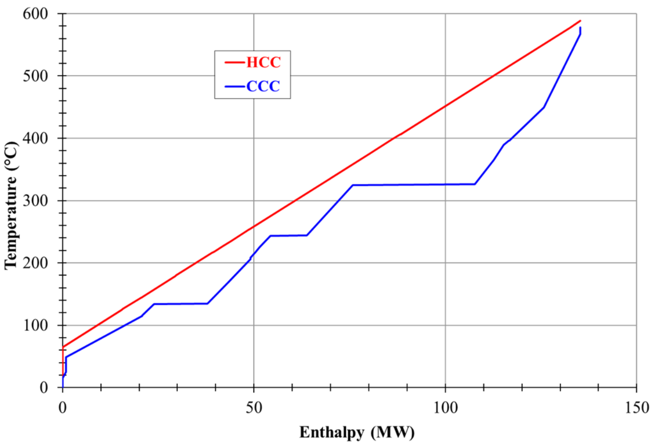

Figure 2. Hot and cold composite curves for decarbonized power generation

As can be noticed from above composite curves, there is no need for external heating utility, the available hot streams within the plant covering the heating duty. Also, one can notice the tight thermal integration which leads to the overall energy optimization within the plant.

The mass and energy balances of optimized systems were then used to quantify the main technical and environmental performances. As benchmark case, the non-carbon capture gasification-based power plant was also considered to evaluate the CO_2 capture energy penalty. The most important performance indexes are presented below:

- Overall energy generation efficiency (η_{Energy}) was calculated considering the global energy output (net power output and energy carrier thermal output) in the overall energy yield of the concepts:

$$\eta_{Energy} = \frac{Net\ power\ output + Energy\ carrier\ termal\ output}{Coal\ thermal\ input} * 100 \quad (6)$$

- Carbon capture rate (CCR) was calculated considering the molar fraction of carbon feedstock that was captured in the Acid Gas Removal unit:

$$CCR = \frac{Captured\ CO_2\ molar\ flow}{Input\ carbon\ molar\ flow} * 100 \quad (7)$$

- Specific CO_2 emissions (SE_{CO_2}) was calculated as emitted CO_2 mass flow for each MW of net energy (net power and energy carrier thermal output) output:

$$SE_{CO_2} = \frac{Emitted\ CO_2\ mass\ flow}{Net\ power\ output + Energy\ carrier\ termal\ output} * 100 \quad (8)$$

RESULTS AND DISCUSSION

The first operation scenario of gasification plant was for power generation only. The following case studies were considered:

Case 1: Coal-based gasification power plant without carbon capture;

Case 2: Coal-based gasification power plant with pre-combustion carbon capture using chemical scrubbing (MDEA);

Case 3: Coal-based gasification power plant with pre-combustion carbon capture using physical scrubbing (Selexol^{TM}).

Table 2 shows the most important technical and environmental performance indicators for the above cases.

Table 2. Technical and environmental indicators for gasification power plants

Performance indicator	UM	Case 1	Case 2	Case 3
Coal flowrate	t/h	30.54	38.74	37.89
Coal LHV	MJ/kg	25.35		
Coal thermal energy	MW_{th}	215.05	272.85	266.80
Gross power output	MW_e	115.45	126.10	125.16
Ancillary power consumption	MW_e	15.45	26.10	25.16
Net power output	MW_e	100.00	100.00	100.00
Net power efficiency	%	46.50	36.65	37.48
Carbon capture rate	%	0.00	90.00	90.00
Specific CO_2 emission	kg/MWh	745.10	85.81	84.21

As noticed from Table 2, there is an important energy penalty when decarbonization process is integrated in the gasification-based power plant. The decarbonization energy penalty is about 9.85 net efficiency percentage points for chemical scrubbing and about 9.02 net efficiency percentage points for physical scrubbing. The main reason for the higher energy penalty in case of chemical gas-liquid absorption represents the heat duty for solvent regeneration, which is about 0.65 – 0.8 GJ/t in case of pre-combustion capture (as evaluated in this work) and about 3 GJ/t in case of post-combustion capture [22]. The lower power generation efficiency in case of decarbonized concepts has another negative consequence which is the increasing fuel requirements for the same net power output. In addition, the power generation costs for decarbonized gasification plants are also increasing on average by about 30-40% [23].

The positive consequence of decarbonization is the significant reduction of specific CO_2 emissions in comparison to the benchmark case without carbon capture (Case 1). This key element could enable the further utilization of fossil fuels (in decarbonized plants) in the future, even if the environmental constraints are getting stricter.

To illustrate the influence of CO_2 capture solvent on hot and cold energy utility consumptions as well as for the overall power plant efficiency, Table 3 presents such an analysis for one chemical solvent (MDEA) and two physical solvent (Selexol™ and Rectisol®). One can notice the overall benefits of physical absorption over chemical one for pre-combustion cases.

Table 3. Influence of solvent selection on heating and cooling utility consumptions

Performance indicator	UM	MDEA	Selexol™	Rectisol®
Power consumption	kWh/kg	0.09	0.11	0.12
Heating consumption	MJ/kg	0.65	0.22	0.38
Cooling consumption	MJ/kg	3.30	0.56	0.62
Overall net power efficiency	%	36.65	37.48	37.01

Concluding, one can notice that physical solvents require less heating and cooling utility consumptions but higher power consumption for solvent circulation [24]. On the other hand, chemical solvents are more selective when various acid gas components are present in gas stream to be treated.

The next evaluated operational scenario of gasification-based plants was based on flexible energy vector poly-generation as a key element for improving plant cycling. Improved cycling capabilities of fossil-based plants is a fundamental important aspect of modern energy conversion systems which need to integrate more time-irregular renewable sources such as solar and wind [13].

The first evaluated scenario refers to the hydrogen and power co-generation based on decarbonized gasification concept. In this design, a variable share of hydrogen-rich stream (after Acid Gas Removal unit) is not sent to the combined cycle for power generation but it is purified in a Pressure Swing adsorption (PSA) unit to purities suitable for external customers (e.g., chemical applications, hydrogen-driven transport). In this way, the overall plant cycling capability (the ability of the plant to timely change the generated energy vectors in accordance to grid demand) are improved.

To illustrate the influence of flexible hydrogen output on overall plant performances, Table 4 presents the case of coal-based Shell gasification plant equipped with Selexol™-based decarbonization unit (Case 3).

If a fully flexible hydrogen and power co-generation plant is targeted, a separate steam cycle has to be used to cover the ancillary power consumption of the plant [25]. This separate power block will use advantages of existing steam-rising capabilities within the plant (e.g., exothermic chemical reactions such as water gas shift, synthetic fuels reactors). The combined cycle is then used only for export power. To illustrate how the overall energy efficiency is varying in case of modification of operation scenario from only power generation to only hydrogen production, Figure 3 presents the situation in case of coal-based Shell gasification plant equipped with Selexol™-based decarbonization unit (Case 3).

Table 4. Performances of decarbonized hydrogen and power co-production

Performance indicator	UM	Power only	Hydrogen and power	
Coal flowrate	t/h	37.89		
Coal LHV	MJ/kg	25.35		
Coal thermal energy	MW_{th}	266.80		
Gross power output	MW_e	125.16	109.91	94.89
Hydrogen thermal output	MW_{th}	0.00	25.00	50.00
Ancillary power consumption	MW_e	25.16	25.51	25.94
Net power output	MW_e	100.00	84.40	68.95
Net power efficiency	%	37.48	31.63	25.84
Hydrogen thermal efficiency	%	0.00	9.37	18.74
Overall plant efficiency	%	37.48	41.00	44.58
Carbon capture rate	%	90.00	90.00	90.00
Specific CO_2 emission	kg/MWh	84.21	76.97	70.79

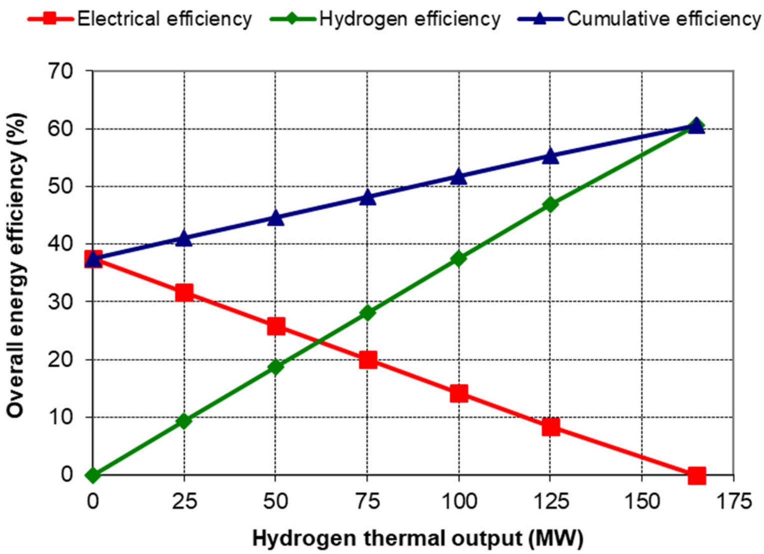

Figure 3. Variation of overall plant energy efficiency vs. hydrogen thermal output

As can be observed from Table 4 and Figure 3, the overall cumulative energy efficiency is favourably influenced by increasing the hydrogen thermal output. Also, the specific CO_2 emission is decreasing with hydrogen thermal output. In addition, fully flexible decarbonized co-generation plants can have a high overall energy efficiency (up to 60%). Accordingly, the flexible hydrogen and power co-generation is a promising operational scenario to improve the gasification plant cycling.

Next evaluated operational scenario for decarbonized gasification plants was energy vector poly-generation based on syngas processing [9]. As evaluated energy vectors (beside electricity), various chemical species were considered as follow: substitute natural gas (SNG), methanol and Fischer-Tropsch fuel. The assessed concepts were designed to be self-sustainable in term of power (available heat sources from various process streams and reactors are used to generate steam which then is converted to electricity in a single cycle power block). All these poly-generation designs consider the once-trough configuration in which the unreacted chemical species from synthetic fuel step are then used for power generation. In this way, the overall flexibility of the plant is improved as well as reducing the design complexity [26].

Table 5 presents the main technical and environmental performances of coal-based Shell gasification plant equipped with Selexol™-based decarbonization unit (Case 3).

Table 5. Performances of decarbonized energy vector poly-generation

Performance indicator	UM	SNG	MeOH	FT fuel
Coal flowrate	t/h	21.93	28.36	25.06
Coal LHV	MJ/kg	25.35		
Coal thermal energy	MW_{th}	154.43	199.71	176.50
Gross power output	MW_e	20.40	26.07	27.42
SNG thermal output	MW_{th}	100.00	-	-
Methanol thermal output	MW_{th}	-	100.00	-
FT fuel thermal output	MW_{th}	-	-	100.00
Ancillary power consumption	MW_e	14.41	15.41	10.69
Net power output	MW_e	5.99	10.66	16.73
Net power efficiency	%	3.87	5.34	9.47
Carrier thermal efficiency	%	64.75	50.07	56.65
Overall plant efficiency	%	68.62	55.41	66.12
Carbon capture rate	%	60.12	48.25	47.62
Specific CO_2 emission	kg/MWh	6.98	26.01	39.85

The first important conclusion regarding the partial decarbonized energy vector poly-generation based on syngas processing is that the overall energy efficiency is significantly higher (55 - 68% vs. 37 - 44%) in comparison to power and hydrogen co-production (fully decarbonized energy carriers). This positive result comes however with a lower carbon capture rate (48 – 60% vs. 90%) which, in the end, means higher CO_2 emissions based on whole life cycle assessment (including energy carriers' usage).

In addition, several important elements can be concluded from the results such as: the combination of chemical synthesis with conventional heat and power production induces a higher overall energy efficiency of the system; once-trough poly-generation concepts have more technical and environmental benefits that recycled plants and decarbonized poly-generation plants have lower energy and cost penalties for carbon capture than conventional stand-alone gasification power plants (operated in base load conditions) [27-28].

CONCLUSIONS

The integration of carbon capture and utilization feature into gasification process for flexible energy vectors poly-generation is assessed in the present paper considering various technical and environmental performance indicators. Both total (power and hydrogen) and partial (synthetic natural gas, methanol, FT fuel) decarbonized energy carriers were considered. Two commercial pre-combustion carbon capture options based on chemical and physical gas-liquid absorption were evaluated. For power generation only, the carbon capture energy penalty was about 9 net percentage points for physical absorption and about 9.8 percentage points for chemical absorption. The main explanation for this fact is that physical gas-liquid absorption using SelexolTM has lower ancillary heat duty for solvent regeneration than MDEA-based scrubbing (0.22 vs. 0.65 MJ/kg).

For flexible energy vectors poly-generation, several important conclusions were drawn. For instance, in case of hydrogen and power co-generation, the overall plant energy efficiency is increasing with hydrogen output (about 3.5 percentage points per each 25 MW_{th} hydrogen output). In case of partial decarbonized energy carriers, the overall energy efficiency is higher (55 – 68%) but carbon capture rate is lower (48-60 vs. 90%) than for total decarbonized energy carriers (power and hydrogen). The overall conclusion is that the decarbonized gasification plant has promising potential for flexible energy vectors poly-generation.

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