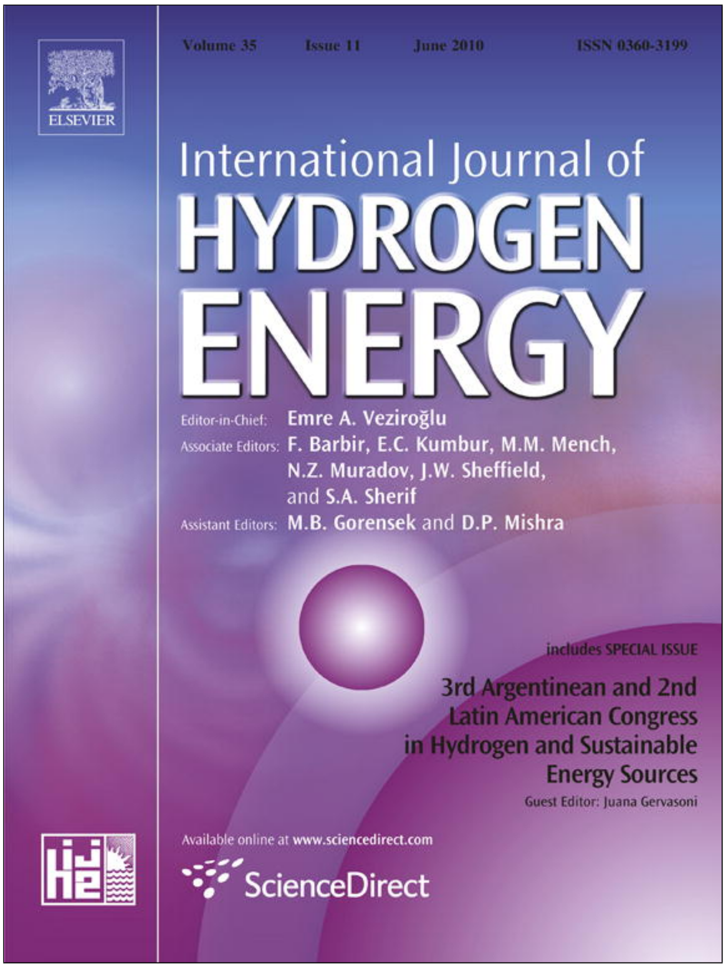
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Recycling a hydrogen rich residual stream to the power and steam plant

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ABSTRACT

The benefits of using a residual hydrogen rich stream as a clean combustion fuel in order to reduce Carbon dioxide emissions and cost is quantified. A residual stream containing 86% of hydrogen, coming from the top of the demethanizer column of the cryogenic separation sector of an ethylene plant, is recycled to be mixed with natural gas and burned in the boilers of the utility plant to generate high pressure steam and power. The main advantage is due to the fact that the hydrogen rich residual gas has a higher heating value and less CO_2 combustion emissions than the natural gas. The residual gas flowrate to be recycled is selected optimally together with other continuous and binary operating variables. A Mixed Integer Non Linear Programming problem is formulated in GAMS to select the operating conditions to minimize life cycle CO_2 emissions.

1. Introduction

The use of internal process streams containing hydrogen is being analyzed industrially. The objectives of a process designer are to maximize the amount of mass ending as final product, thereby minimizing wastes, emissions streams, energy and operating cost. Fu and Diwekar [1] and Li et al. [2] have proposed the minimization of cost and gas emissions with a multi objective framework for utility plants, while the former authors have worked with NOx and SOx emissions, the later have worked with GHG emissions. Li et al. [2] also considered the life cycle emissions associated with natural gas used as fuel in the utility plant. Corrado et al. [3] present the environmental assessment of a steam and power plant that burn H_2 as fuel.

The present work presents a methodology to optimize the mass and energy integration of an ethylene production plant with the steam and power sector. The operating conditions of a steam and power plant are optimized, including the flowrate of a hydrogen rich residual gas stream recycled from the ethylene plant. The ethylene is produced from ethane. A superstructure optimization is formulated to select the utility plant operating conditions minimizing the life cycle CO_2 emissions. To assess life cycle CO_2 emissions the battery limits of the steam and power sector are extended to include not only the combustion emissions but also the main sources of CO_2 emissions corresponding to natural gas and imported electricity generated by thermoelectric, hydroelectric and nuclear plants. The operating conditions to be selected in utility systems are: hydrogen rich residual gas recycled, temperature and pressures of high, medium and low pressure steam headers, deaerator pressure and letdowns flow rates. Binary operating variables represent discrete decision such as equipment that can be on or off as boilers and their auxiliary

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Nomenclature

e	CO_2 emission factor
E	Energy content
F	Stream flow rate
H	Specific enthalpy
LHV	Lower heating value
Q	CO_2 emissions flowrate
x	Energy contribution fraction

Superscripts

LC	Life cycle

Subscripts

i	Utility plant stream
IE	Imported electricity
j	fuel stream: NG or RG
NG	Natural gas
RG	Residual gas
Tot	Total
UP	Utility plant

Unit sets

EM	Electrical motors
ST	Steam turbines

equipment and the selection between optional drivers for pumps that can be either electrical motors or steam turbines. The numerical results show significant reductions in CO_2 emissions, natural gas, water and electricity consumption and therefore operating cost. The electrical motors are switched to steam turbines because the latter have a smaller ratio of life cycle CO_2 emissions to power generated than in the former, due to the fact that residual gas, rich in hydrogen, is burned with natural gas reducing the CO_2 emissions of the steam and power sector.

2. Chemical process description

Ethylene is produced from ethane. Natural gas is mixed with residual gas from the demethanizer column and burned in the pyrolysis furnaces. The ethane is heated with a steam mix in pyrolysis furnaces, a process which breaks down or cracks ethane into ethylene, hydrogen and other by products. Sudden cooling then stops the reaction and the subsequent mixture of gases is compressed, cooled and separated in a series of distillation towers. The first cryogenic distillation tower, called demethanizer, has a top product stream composed mainly by hydrogen and at less extend by methane. This hydrogen rich stream is recycled and re-used for steam generation in the utility plant and in the furnaces. The entire processes consist of eight pyrolysis furnaces, a cracked-gas compressor, heat recovery network, a cryogenic separation sector with eight distillation towers, the refrigeration system and the steam generation system. Sudden cooling that stops the reaction is done with water that simultaneously generates high pressure steam in the quenching sector. A schematic flow sheet of the ethylene process is shown in Fig. 1, below.

A residual stream from the top of the demethanizer column that contains mainly hydrogen and at less extent methane is recycled and mixed with natural gas to be burned in the boiler of the utility plant and in the cracking furnaces to generate steam and power. The residual stream is shown in doted lines in Fig. 1.

3. Steam and power plant

The utility plant provides mainly steam and power to the chemical plant. It consumes fossil fuels, a non-renewable resource burnt in the boilers and also a scarce resource as water. A schematic flow sheet of the utility sector of an ethylene plant is presented in Fig. 2.

There are four boilers B1 to B4 and a waste heat boiler, associated with the quenching sector of the ethane cracking furnace, that produce superheated steam at high pressure (HPS). The main equipment of the utility plant superstructure are: high (HPSH), medium (MPSH) and low (LPSH) pressure steam headers, steam turbines, pumps, deaerator tank, vents, letdown streams, water treatment plant, vacuum condenser tank, air fans, electrical motors, heat exchangers, etc. The utility plant model includes continuous operating variables such as residual gas flowrate, temperature and pressure of the high, medium and low pressure steam headers and deaerator pressure. The modeling equations of the main equipment and steam and water enthalpy and entropy property predictions are posed as equality constraints in GAMS [4]. The plant has alternative drivers such as electrical motors (EM) and steam turbines (ST) for some pumps. Binary variables are used for the selection of alternative drivers and some other equipment that can be on or off, like boilers and their auxiliary equipment (pumps and air fans). The main power demands of the process plant correspond to three compressors: cracking gas (CGC), propylene gas (PC) and ethylene gas (EC). The condensed steam returning from the chemical process heat exchangers is collected in a tank at atmospheric pressure to be re-used and generate steam. The recycled condensate cannot be re-used without previous water treatment to prevent corrosion in boilers and steam turbines. There are optional drivers that can be electrical motors (M1 to M11) or steam turbines (T1 to T11) for eleven pumps in Fig. 2. A detailed mathematical model of the utility plant is presented in Eliceche et al. [5].

3.1. *Estimation of combustion CO_2 emissions of the utility plant*

Both, boilers and waste heat boiler consume a mixture of natural gas and residual gas recycled from the ethylene plant. The CO_2 emissions during combustion of natural and residual gas are quantified using stoichometric emission factors. The stoichiometric emission factors are calculated with the chemical composition of each fuel gas following Davis [6] and IPCC [7]. The natural gas burned in the utility plant has a weighted average composition as a result of natural gases from different country pipelines changing along the year, having 90% of CH_4 and 2% of CO_2. The residual gas comes from the top of the demethanizer column in the ethylene plant and

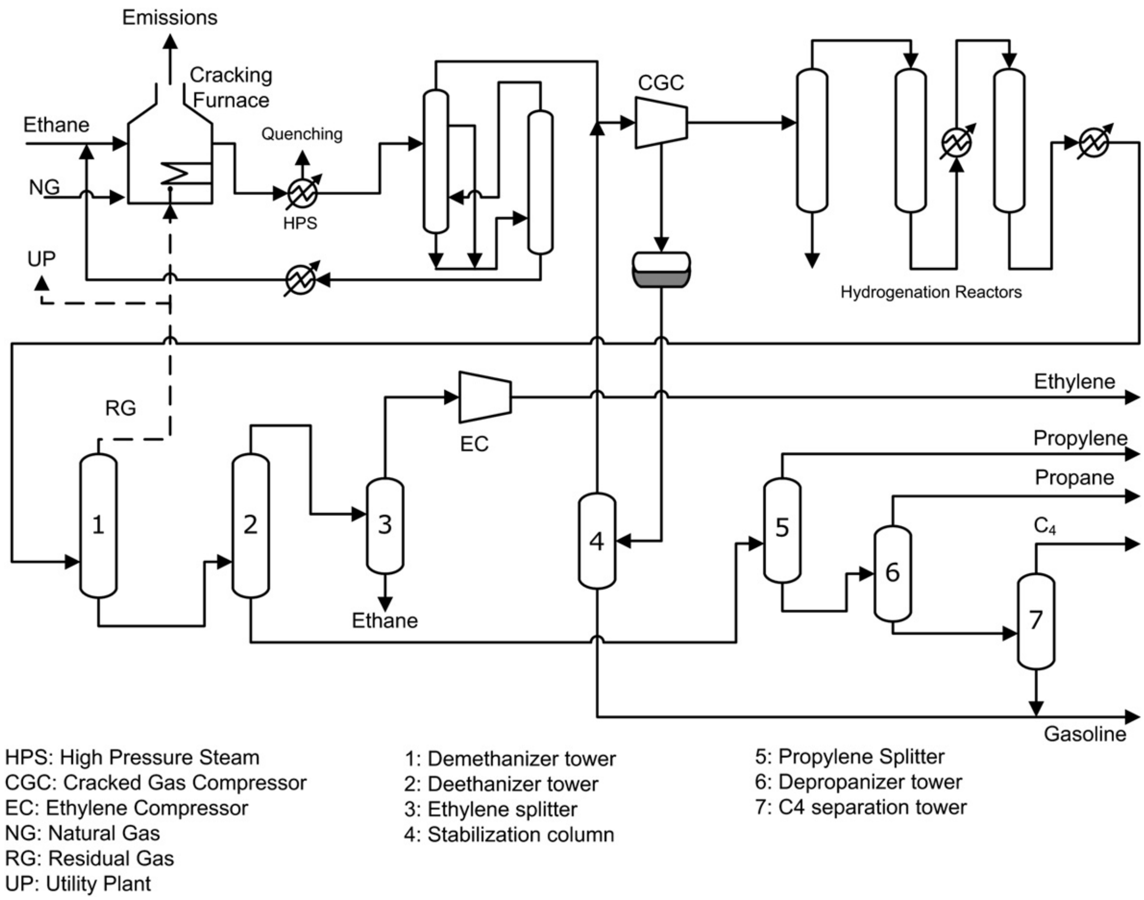

Fig. 1 – Ethylene process flowsheet.

it is composed by H_2 86% and CH_4 14%. Thus the CO_2 emission from the utility plant Q_{UP}, due to the combustion of natural and residual gases is calculated as follow:

$$Q_{UP} = F_{NG} \times e_{NG} + F_{RG} \times e_{RG} \tag{1}$$

Where F_j represents the mass flow rate of fuel stream j and e_j is the CO_2 combustion emission factor for fuel stream j expressed in [mass of CO_2/mass of fuel gas], for the sake of simplicity the subscript "CO_2" in the emission factors has been omitted. The lower heating values (LHV) of each gas, as well as the CO_2 emission factors are presented in Table 1.

As can be seen in the previous table, the residual gas stream has a higher lower heating value and a lower CO_2 emission factor than the natural gas. Thus the ratio of CO_2 emissions to power delivered is lower for the residual gas than for the natural gas as shown in Table 1.

3.2. Estimation of natural gas life cycle CO_2 emissions

Natural gas life cycle CO_2 emissions sources are mainly flaring combustion at the gas well and leakages during transportation. The combustion emissions of the vented natural gas at the well are considered considering that almost 2% of the gas well production is flared, National Greenhouse Gases Inventory [8]. The leakages during transportation are produced in the compressor stations, where the natural gas pressure is adjusted for distribution, and a small quantity of natural gas is lost in the pipeline itself due to minor failures in the pipeline material as corrosion, Natural Gas Organization [9]. Therefore, the life cycle stages considered for natural gas life cycle are: extraction, production and transportation. The CO_2 emissions during the natural gas life cycle Q_{NG}^{LC} are added to the generation in the utility plant given in Eq. (1).

3.3. Imported electricity life cycle CO_2 emissions quantification

There are electrical motors that consume electricity, imported from the national interconnected electricity network, generated by thermoelectric, hydroelectric and nuclear power plants.

In the life cycle approach it is required to analyze the CO_2 emissions which arise upstream and downstream of the power plant, otherwise the CO_2 emissions resulting from electricity generation of the various fuel options are underestimated as stated by Weisser [10]. Heavy metals, particulate matter and radioactive emissions produce significant environmental and health impact in the electricity generation supply chain, IEA [11], although they are not considered in this work.

Each electricity generation option has CO_2 emissions associated with its life cycle including extraction of raw materials as petroleum, natural gas and uranium, the generation step itself and waste disposal. Natural gas, fuel oil and gas oil are used in thermoelectric power plants. The life cycle stages considered are: raw material extraction, transport,

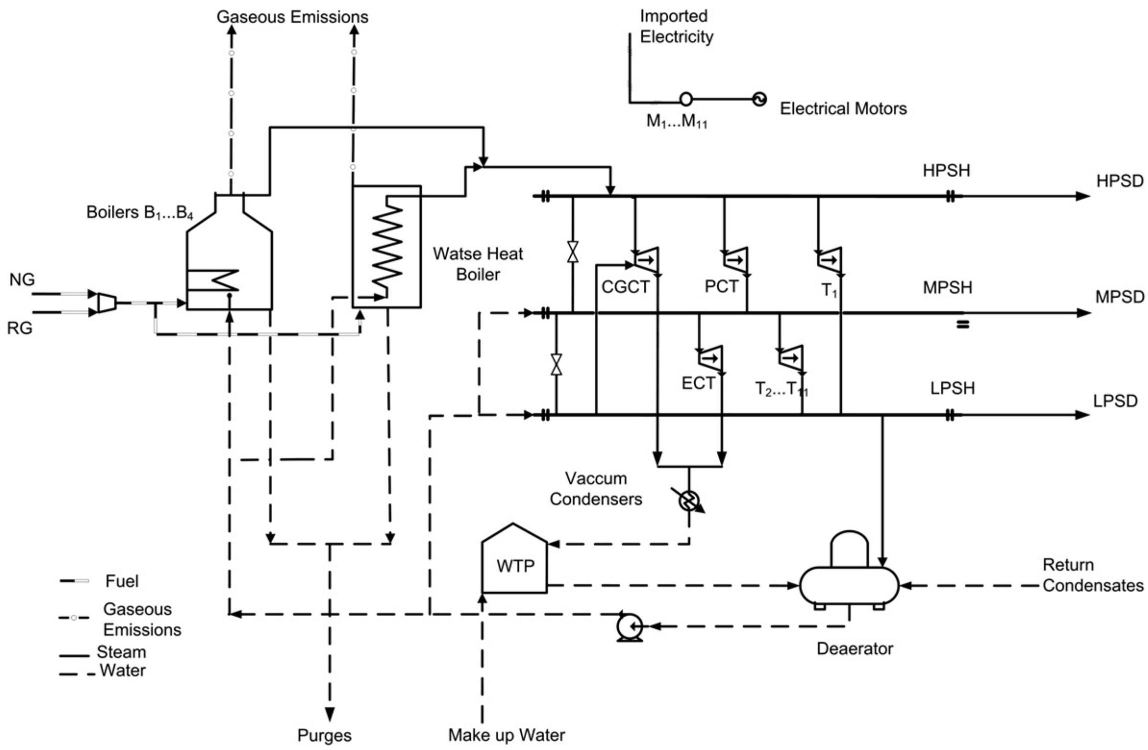

Fig. 2 – Steam and power plant flowsheet.

refining and electricity generation itself. The life cycle stages considered in hydroelectric power generation are: the dam construction, generation and the biomass decay during the power station operational phase. For nuclear power generation the following life cycle stages of the uranium fuel are considered: mining and milling, transport, conversion and enrichment stages, fuel rod assembly, electricity generation, spent fuel waste storage and the construction stage of nuclear power plants. Then, the life cycle CO_2 emissions of the imported electricity, Q_{IE}^{LC}, are quantified adding the CO_2 emissions of all the life cycle stages of thermoelectric, hydroelectric and nuclear electricity imported.

4. Selection of the operating conditions of the utility plant and hydrogen rich residual gas recycled

The selection of the hydrogen rich residual flowrate recycled from the ethylene plant to the utility sector, the temperature and pressure of the steam headers and the selection of alternative drivers and the boilers in operation is carried out minimizing the life cycle CO_2 emissions calculated in the following section.

4.1. *Objective function*

The objective function is the overall life cycle CO_2 emissions flow rate Q_{Tot}^{LC}, which is calculated adding the utility plant life cycle CO_2 emissions, natural gas life cycle CO_2 emissions Q_{NG}^{LC} and imported electricity life cycle CO_2 emissions Q_{IE}^{LC},

$$Q_{Tot}^{LC} = Q_{UP} + Q_{NG}^{LC} + Q_{IE}^{LC} \tag{2}$$

4.2. *Optimization problem formulation*

The following optimization problem is formulated to select the hydrogen rich residual gas flowrate recycled from the ethylene plant and the optimal operating conditions of the steam and power plant to minimize the overall life cycle CO_2 emissions flow rate Q_{Tot}^{LC} as it was defined in Eq. (2). A Mixed Integer Non Linear Programming problem (P1) is formulated and solved in GAMS [4].

$$\begin{aligned} \min_{x,y} \quad & Q_{Tot}^{LC}(x,y) \\ \text{s.t.}: \quad & h(x,y) = 0 \\ & g(x,y) \le 0 \\ & x^L \le x \le x^U \\ & x \in R^n \\ & y \in \{0.1\}^m \end{aligned} \tag{P1}$$

Where x and y are continuous and binary variables. Superscripts U and L indicates upper and lower bounds on the

Table 1 – Lower heating values, CO_2 emissions and CO_2 emissions to power ratios.

Fuel stream	*LHV* (kWh/ton)	CO_2 emission factor (kg CO_2/ton)	CO_2 emission to power ratio (kg CO_2/kWh)
Natural gas	13 571.356	2703.546	0.1992
Residual gas	22 304.529	1566.612	0.0702

Table 2 – Improvements achieved minimizing life cycle CO_2 emissions.

		Initial point	Optimal solution	Reduction %
Residual gas	ton/h	3.00	4.96	−39.52
Life cycle CO_2 emissions	kg/h	37 835.02	27 867.89	26.34
Utility plant CO_2 emissions	kg/h	36 110.38	27 146.36	24.82
UP natural gas life cycle CO_2	kg/h	465.47	287.15	38.31
Imp. Elect. CO_2 emissions	kg/h	1259.17	434.37	65.50
Operating cost	$/h	2733.08	1638.35	40.05
Natural gas	ton/h	11.62	7.17	38.29
Make up water	ton/h	32.00	22.00	31.25
Boilers high pressure steam	ton/h	87.18	68.19	21.78
Cracking furnace high pressure steam	ton/h	104.18	101.97	2.12
Imported electricity	kWh	3114.58	1074.42	65.50

continuous variables x. Hydrogen reach residual gas flowrate, pressures and temperatures of high, medium and low-pressure steam headers, deaerator pressure and letdowns are the continuous operating variables, a subset of vector x. Binary variables y represent discrete decisions that allow the selection of alternative pumps drivers such as steam turbines and electrical motors and whether the boilers and their corresponding feed pumps and air fans are on or off.

Equality constraints $h(x,y) = 0$ represent the steady state modeling of the steam and power generation plant, including mass, energy balances and steam properties prediction. Inequality constraints $g(x,y) \leq 0$ represent: logical constraints, minimum and maximum equipment capacities, operating and design constraints, etc. The power and steam demands of the ethylene plant are posed as equality constraints. The main power demands correspond to the cracked gas, ethylene and propylene refrigeration system compressors. Other power demands correspond to condensate pumps, boiler water pumps, cooling water pumps and air fans.

Binary variables are associated to the selection of alternative pumps drivers and equipment that can be on or off. Electrical motors and steam turbines are optional drivers for lubricating pumps, condensate pumps, boiler water pumps and cooling water pumps. Boilers and their corresponding air fans and pumps can be on or off. A detailed model of the system of equations including binary variables has been presented by Eliceche et al. [5].

5. Numerical results

The hydrogen rich residual gas coming from the top of the demethanizer column of the cryogenic separation sector of an ethylene plant obtained from ethane is recycled and mixed with natural gas to be burned in the boilers of the utility plant. The operating conditions of the utility plant are selected optimally to minimize life cycle CO_2 emissions solving the MINLP problem P1 in GAMS [4]. The MINLP problem was solved with DICOPT as the outer approximation option, CONOPT3 was used to solve the nonlinear programming sub problem and CPLEX was used to solve the mixed integer linear programming sub problem. The upper bound on availability of residual gas to be recycled is 4.96 ton/h. The main improvements achieved with the selection of the operating conditions solving the optimization problem (P1) are presented in Table 2.

A reduction in the order of 26% in life cycle CO_2 emissions is achieved solving the MINLP problem P1. Simultaneously a reduction of 38% is observed in natural gas consumption, the non-renewable fossil fuel burned in the boilers, leading to a reduction of 40% in the operating cost of the utility sector. Operating cost is reported in Argentinian pesos, the exchange rate being equal to 1 USA = 3.8 $. Thus, the minimization of life cycle CO_2 emissions has led to significant reduction in operating cost and natural gas consumption due to a more energy efficient operation that can be achieve though process optimization.

5.1. *Selection of optimal flowrate of the recycled residual gas, temperature and pressure of steam headers*

Residual gas flowrate recycled, temperature and pressure of high, medium and low pressure steam headers are calculated to minimize the life cycle CO_2 emissions and their values are reported in Table 3. The imported electricity scenario considered correspond to: 53% of thermoelectric (87% natural gas and 13% oil), 35% of hydroelectric and 12% of nuclear

Table 3 – Hydrogen rich recycle, temperature and pressures of steam headers.

Optimization variables		Initial point	Solution point	Lower bound	Upper bound
Residual gas	ton/h	3.00	4.96	0.00	4.96
HPSH temperature	°C	420.00	450.00	400.00	450.00
MPSH temperature	°C	320.00	310.00	310.00	370.00
LPSH temperature	°C	210.00	150.00	150.00	250.00
HPSH pressure	bar	50.50	52.00	48.00	52.00
MPSH pressure	bar	23.00	23.81	18.00	26.00
LPSH pressure	bar	3.00	4.01	3.00	5.00
Deaerator Pressure	bar	2.50	2.67	1.20	3.00

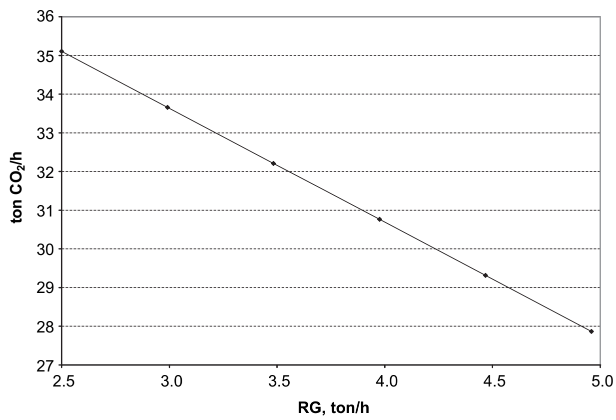

Fig. 3 – Life cycle CO_2 emissions vs hydrogen residual gas flowrate.

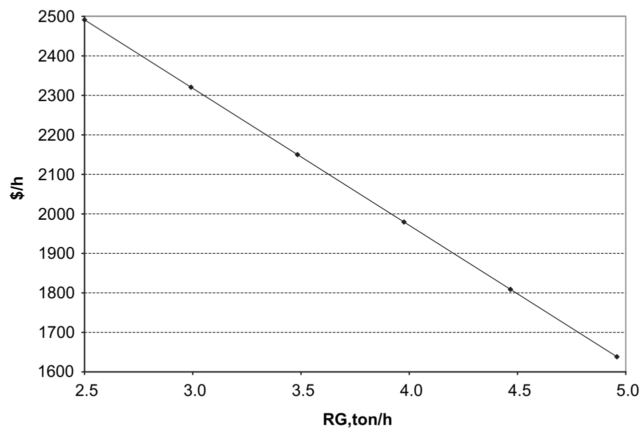

Fig. 4 – Operating cost vs hydrogen residual gas flowrate.

generation. Residual gas flowrate, temperature and pressure of high pressure steam header reach their upper bounds values while temperature of low pressure steam header reaches its lower bound to increase the power that can be delivered, increasing the process efficiency and the ratio of power delivered with respect to natural gas burned in boilers. To minimize the CO_2 emissions, the natural gas consumption is minimized and the residual gas recycled is maximized. So a proper selection of temperature and pressure of steam headers is crucial to reduce natural gas consumption and consequently the CO_2 emissions.

The sensitivity of life cycle CO_2 emissions with respect to the residual gas flowrate is shown in Fig. 3 and the sensitivity of the operating cost is shown in Fig. 4. Both functions are monotonic decreasing linearly. Furthermore, a reduction of 2.9 ton of life cycle CO_2 emissions per ton of residual gas recycled can be estimated indicating the relevance of this strategy of mass integrating between the ethylene plant and the steam and power sector. In terms of operating cost reduction a ratio of 356 $ per ton of residual gas recycled can be expected. These two ratios can support a decision making process, quantifying the benefits of recycling the residual gas in terms of life cycle CO_2 emissions and operating cost reductions.

A reduction of 34.5% in operating cost is due to the increment of residual gas recycled from 2.5 ton to 4.9 ton.

5.2. *Discrete decisions represented by binary variables*

For a given configuration of alternative pump drivers the selection between steam turbines (ST) and electrical motors (EM) is carried out using binary optimization variables. Binary variables associated with a given driver have values of 1 or 0: if the value assigned solving problem (P1) is 1, the optional driver has been selected and it is on; if the value assigned solving problem (P1) is 0, the optional driver has not been selected and it is off. Binary variables are also used to select which boilers and their auxiliary equipment (such as pumps and air fans) are on or off. The main results concerning the binary optimization variables are presented in Table 4.

There are twenty-four binary variables. Twelve of the binary variables are used to select between electrical motors and steam turbines as drivers. The rest of the binary variables are used to define if the equipment is ON or OFF. At the initial point of problem (P1) there are three boilers ON and at the solution point there are only two boilers ON, thus the air fan corresponding to boiler 3 and 4 are OFF. Ten electrical motors at the initial point of problem (P1) are switched to steam turbines at the solution point. One electrical motor that was ON at the initial point is turned off at the solution point. Finally, one steam turbine corresponding to a cooling water pump that was ON at the initial point is turned off at the solution point. At the optimal point, electrical motors are switched to steam turbines because the latter have a smaller ratio of CO_2 emissions to power generated, due to the fact that the residual gas, rich in hydrogen, is burned with natural gas reducing the CO_2 emissions.

6. Analysis of the ratio of CO_2 emissions to power generated

The CO_2 combustion emissions allocation is made accounting the energy contribution of each output stream leaving the utility plant with respect to the total energy obtained by the natural and residual gas combustion. The energy generated by natural and residual gas combustion is calculated adding the combustion energy liberated by each gas multiplying each gas mass flow rate by its lower heating value, as follows:

Table 4 – Selection of alternative drivers and boilers.

		Initial point	Solution point
Steam turbines	ON	2	11
Electrical motors	ON	13	2
Boilers	ON	3	2
Steam turbines	kWh	2496.75	1950.353
Electrical motors	kWh	940.810	988.466
Total power	kWh	3887.56	2938.819

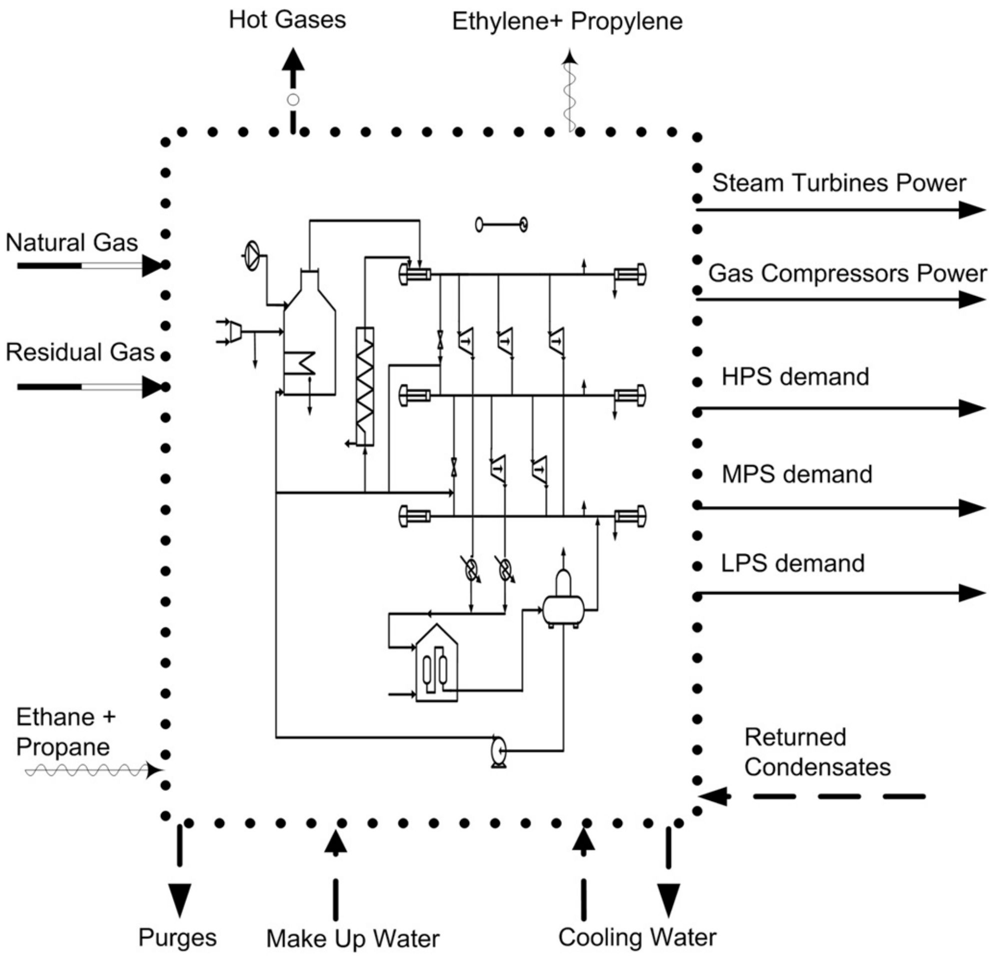

Fig. 5 – Utility plant main energy streams.

$$E_C = F_{NG} \times LHV_{NG} + F_{RG} \times LHV_{RG} \quad (3)$$

Where E_c is the energy generated by the natural and residual gas mixture combustion, F_j is the mass flowrate of the fuel gas j and LHV_j is the lower heating value of fuel gas j calculated with the corresponding composition and expressed in [kWh/mass unit].

The energy content of each stream can be calculated as follows:

$$E_i = F_i \times (H_i - H_{ref}) \quad (4)$$

Where E_i is the energy content of each output stream i and F_i is the mass flow rate of stream i, H_i is the specific enthalpy of the stream i, and H_{ref} is the specific enthalpy of the returned condensates, at 98 °C and 1 atm. The specific enthalpy of each stream is calculated with an appropriate enthalpy and entropy property prediction included in the modeling of the utility as shown in Eliceche et al. [5].

The ratio between the energy content of each stream and the total energy produced by the fuel mix combustion is defined in Eq. (5) as x_i,

$$x_i = \frac{E_i}{E_C} \quad (5)$$

The CO_2 emissions allocation methodology proposes that the ratio between the assigned CO_2 emissions of each stream Q_i and the total CO_2 emissions from the combustion of natural and residual gases Q_{UP} is equal to x_i defined previously in Eq. (5),

$$\frac{Q_i}{Q_{UP}} = x_i \quad (6)$$

Q_{UP} is calculated from Eq. (1) and Q_i is calculated from Eq. (6). The main energy streams are shown in Fig. 5 and the CO_2 emissions allocation at the solution point is shown in Table 5.

The energy content of the inlet make up water stream as well as the ethane-propane inlet stream is considered as negligible. The energy input from the fuel mix combustion is 207 880.168 kWh and the total amount of CO_2 emissions is 27 146.36 Kg CO_2. The results reached with the data presented in the previous figure and applying Eq. (6) are summarized in Table 5.

Using the data in the previous table it is possible to calculate the CO_2 to power delivered ratio simply dividing the values in the third column by the values in the second column. Then the ratio is equal to: 0.13 Kg. CO_2/kWh for all the output streams of the steam and power plant.

The ratio of CO_2 emissions to power delivered for imported electricity is calculated dividing the power generated with electrical motors by the amount of CO_2 emissions of the

Table 5 – Utility plant energy and CO_2 emissions allocation.

Output stream	E_i, kWh	Q_i, kg CO_2
Hot gases	13 279.509	1734.138
Ethylene + Propylene	62 902.267	8214.250
HPSD-HPSC	5315.514	694.140
MPSD-MPSC	14 118.928	1843.756
LPSD-LPSC	18 240.624	2381.997
Purges	1367.632	178.596
Cooling water	55 667.383	7269.464
Gas compressors power	25 301.559	3304.067
Steam turbines power	1950.353	254.692
Losses	9734.963	1271.264
Total	207 878.732	27 146.364

imported electricity. This ratio is equal to 0.44 Kg CO_2/kWh. This is the case for the following distribution of electricity power plants: 53% of thermoelectric (87% natural gas and 13% oil), 35% of hydroelectric and 12% of nuclear generation. The ratio of CO_2 emissions to power delivered for the steam turbines in the utility plant is equal to 0.13 Kg CO_2/kWh.

This difference between ratios of CO_2 emissions to power delivered justifies the preference to choose steam turbine drivers instead than electrical motors at the optimal solution point. This fact is an evidence of the advantage of using a hydrogen reach residual gas stream to generate steam and power in the utility sector of a petrochemical process. Moreover, the superstructure optimization increases the efficiency, reducing the CO_2 emissions and operating cost as a result of the process integration between the ethylene plant and the utility sector.

7. Conclusions

The methodology presented is an important tool to support a decision making process in order to use internal hydrogen rich streams to reduce life cycle carbon dioxide emissions and cost, contributing to the fulfillment of the Kyoto protocol and its emission reduction targets. Significant reductions have been achieved not only in CO_2 emissions, but also in natural gas, water and electricity consumption and therefore in operating cost. The recycle and reuse of a hydrogen rich residual gas from the top of a demethanizer column of an ethylene plant to generate steam and power, a key sector in petrochemical plants, is analyzed. A sensitivity analysis of life cycle CO_2 emissions and operating cost with respect to the residual gas flowrate recycled, shown in Figs. 3 and 4, shows the impact of this strategy in reducing the ratio of CO_2 emissions to power delivered and operating cost of the utility plant. The sensitivity analysis is also important in order to compare this combustion option with possible alternative uses of hydrogen and methane of the residual gas stream. It is not strait forward the quantification of Carbon dioxide emissions and operating cost reductions, due to the recycle and reuse of internal process plant streams. The methodology has been applied to analyze the recycle and reuse of an ethylene plant residual gas in the utility sector, although it can be extended to different industrial plants. It is important to extend the battery limits of the process plant to consider life cycle carbon dioxide emissions as it has been done in this work, otherwise misleading results can be obtained.

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