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EXERGY ANALYSIS OF CARBON DIOXIDE CAPTURE USING RENEWABLE SOURCES

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Abstract. *In order to meet the world's energy demands in a cleaner way, the use of renewable energy resources has now been implemented and expanded. However, some of the renewable energies can suffer from problems related to intermittency and seasonality, as with those using solar, water and wind resources. One way to solve generation fluctuations that may not fit the demands is the conversion and storage in other forms of energy, which allows the creation of a reservoir until there is a demand that cannot be supplied by the instantaneous production of energy only. This work aims at calculating the relation between the electric power needed to transform the CO_2 produced on a cement plant on CH_4 and the exergy efficiency of a power-to-methane generation process. The control volume includes electrolysis, methanation, CO_2 capture and the cement production processes. Regarding the methanation, to the CH_4 production it is used the hydrogen provenient from the water electrolysis and the carbon dioxide proceeding from a cement factory. The exergy efficiency found for the whole composition of processes is 48%.*

Keywords: *Exergy, thermodynamics, power-to-gas, energy storage.*

1. INTRODUCTION

The production of electricity from renewable resources increased in the past few years since there is a necessity of reducing non-renewable energy production with CO_2 emissions. Most of the renewable energy production is intermittent, and the peak of production usually is not at the same moment as the demand (Guandalini et al, 2015). Therefore, there are several energy storage systems. The most common methods used are batteries, chemical, physical and thermal (sensible and latent) (Lehner et al, 2014). For each range of energy production, one storage technology is more suitable. Nevertheless, some of them have an environmental impact that must be avoided, for instance batteries that may contaminate the environment and damage the ecosystem on its production and disposal (Zhang et al, 2016).

There are some processes of the cement factories which are responsible to high amounts of CO_2 emissions. This industry is responsible for 5-7% of all CO_2 emitted on earth. The manufactory of one cement tonne produces around 900 kg of CO_2 (Benhelal et al, 2013).

A quocient relating the inputs of energy and CO_2 express the quantity of energy needed from the renewable sources to transform one kilogram of CO_2 on CH_4. An exergy efficiency analysis according to the second law of thermodynamics provides a better understanding of the system compared to ideal conditions of operation (reversible). It takes into consideration the quality of the energy conversions and its degradation during the processes. Therefore, this index gives a quantitative clue of how much of work potential may be lost during the storage process, which is the aim of this work.

2. MATERIALS AND METHODS

2.1 Control Volume

Using the informations that there is a necessity for energy storage that can use CO_2 as input and combined to the fact that the cement industry has some environmental issues regarding pollution, since the process requires the combustion of a very pollutant fuel, it is proposed to combine the CO_2 output from the cement industry to generate methane in a power-to-gas process. This objective can be achieved by incorporating a plant with an electrolysis and methanation processes with CO_2 capture in the output of the cement process. The schematic representation of this plant is indicated in Figure 1.

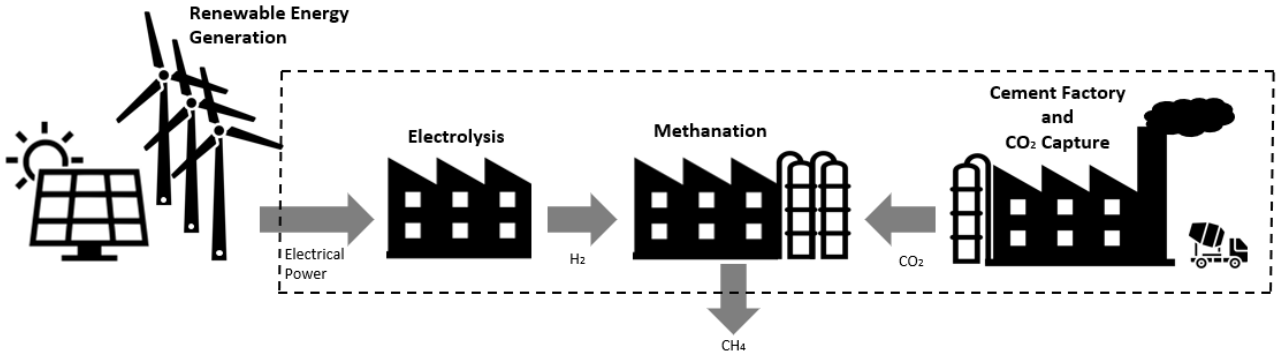

Figure 1. Power-to-methane plant associated with CO_2 captured from a cement factory

The control volume for the exergy analysis is defined by the shaded line in Figure 1. It encompasses the processes of electrolysis, methanation, CO_2 capture and cement production. It must be raised out that the final use by combustion of the CH_4 and its return to CO_2 released to the atmosphere with possible capture is not yet considered in this article.

Regarding the data used, according to the public data provided by J. Mendo consulting, the annual cement production of a single factory in Brazil from the Itambé Group was of 938 thousand tonne of cement in the year of 2007, from its factory located in Balsa Nova, state of Paraná. The CO_2 emissions of the productive process of cement used were taken from the average emissions on the Brazilian scenario, from the report of the Cement Sustainability Initiative "Getting the numbers right", published in 2016, which says that it is produced 572 kg of CO_2 for each tonne of cement. Thus, it was used the value of 5.37 x 10^5 tonnes of CO_2 for one year of cement production and the values of inputs and outputs of the other processes were adapted to fulfill this output/input analysis for the methanation process to have an overview of the electricity needs and to be able to compute the relation between the electricity needed and the CO_2 input.

2.2 Exergy

The exergy analysis provides a value to the analysis of the energy degradation related to the input energy (Tsatsaronis, 1993). The term "exergy" represents the maximum theoretical work that can be obtained from an amount of matter, when it is taken to the thermodynamic equilibrium (mechanical, thermal and chemical) with the environment, involving exclusively interactions with the environment components through reversible processes (Szargut; Morris; Steward; 1988).

In order to properly apply the exergy analysis, the exergy of each stream must be calculated. Equation (1) can be used to assess the physical and chemical exergy of a stream. In Equation (2) the physical exergy was opened as a function of the enthalpy, entropy and the environmental reference (T_0 = 298K, P_0 = 1 atm)

$$b_{total} = b_{flow} + b_{ch} \tag{1}$$

$$b_{flow} = h - h_0 - T_0(s - s_0) \tag{2}$$

Where:

b_{total} : Total Exergy (kJ/kg)
b_{flow} : Specific flow exergy (kJ/kg)
b_{ch} : Chemical Exergy (kJ/kg)
h : Enthalpy (kJ/kg)
h_0 : Enthalpy at environmental reference (kJ/kg)
s : Entropy (kJ/kg K)
s_0 : Entropy at environmental reference (kJ/kg K)

The enthalpy difference and entropy difference can be written as follows in Equations (3) and (4)

$$\Delta h = h - h_0 = \int_{T_0}^{T} c_p(T)\, dT \tag{3}$$

$$\Delta s = s - s_0 = \int_{T_0}^{T} c_p(T)\, \frac{dT}{T} - R \cdot ln\left(\frac{P}{P_0}\right) \tag{4}$$

For a control volume at steady state the destroyed exergy is defined in Equation (5)

$$\dot{B}_d = \sum \dot{m}_{in}\, b_{in} - \sum \dot{m}_{out}\, b_{out} + \dot{Q}\left(1 - \frac{T_0}{T}\right) - \dot{W_{CV}} \tag{5}$$

Where:

$\dot{B}_d$: Destroyed Exergy (kJ/s)
$\dot{m}$: Mass flow (kg/s)
$\dot{b}$: : Exergy flow (kJ/kg)
$\dot{Q}$: Heat exchange (kJ/s)
T_0 : Room temperature (298.2 K)
T : Temperature of heat Exchange (K)
$\dot{W}_{CV}$: Work (kJ/s)

The exergy efficiency can be expressed in terms of the control volume as follows on Equation (6). This definition is usually current in a chemical process where there is no useful effect.

$$\eta_{ex} = \frac{B_{out}}{B_{in}} \tag{6}$$

Where

η_{ex} : Exergy efficiency
B_{out} : Exergy leaving the control volume (kJ/kg)
B_{in} : Exergy entering the control volume (kJ/kg)

2.3 Subprocesses

2.3.1 Cement Production

The cement factories produce cement from physical and chemical processes over limestone and a second material as clay. Clinker is the name given to the product of the burning of limestone and from its mixture with additives (gypsum or anhydrite) it is obtained cement.

The process of cement production occurs as follows: first of all, extracting the raw material from the crust of the earth. Then, this material is taken to the factory, where it passes through mechanical processes of breaking (crushing and milling), and enters the kiln, where it passes through the chemical processes of calcination and sintering that occurs at temperatures of 700 to 800 ºC (Vizcaíno-Andrés et al, 2015; Mujumdara; Ranade, 2006) and 1450 to 1550 ºC, respectively (Rodrigues et al, 2013; Koroneos et al, 2005).

The calcination reaction happens as follows in Equation (7) and (8).

$$CaCO_3 \rightarrow CaO + CO_2 \tag{7}$$

$$MgCO_3 \rightarrow MgO + CO_2 \tag{8}$$

The sintering reaction obeys the Equations (9) to (12).

$$2\,CaO + SiO_2 \rightarrow (CaO)_2SiO_2 \tag{9}$$

$$CaOAl_2O_3 + 2CaO \rightarrow (CaO)_3Al_2O_3 \tag{10}$$

$$CaOAl_2O_3 + 3CaO + Fe_2O_3 \rightarrow (CaO)_4Al_2O_3Fe_2O_3 \tag{11}$$

$$(CaO)_2SiO_2 + CaO \rightarrow (CaO)_3SiO_2 \tag{12}$$

There are several types of cement, depending on the percentages of its composites. The composition used in this study follows the composition of Portland ASTM Cement type I (Mindess et. al, 2002).

To the exergy calculation the relations showed in Table 1 for the composites were used (Rodrigues et. al, 2013), where T is the temperature of the chemical species.

Table 1. Data of each chemical species on clinker constitution

Chemical species	Specific Heat (J/mol.K)	Chemical Exergy (kJ/kg)
CaO	$41.8 + 0.0202 \text{ x T} - (451440)/T^2$	1.7540
SiO_2	$53.504 + 0.0186846 \text{ x T} - (1262360)/T^2$	0.028881
Fe_2O_3	$103.3296 + 0.0670472 \text{ x T} - (1769812)/T^2$	0.12791
$(CaO)_3Al_2O_3$	$60.987872 + 0.02678962 \text{ x T} - (1109831{,}8)/T^2$	6.1849
$CaOAl_2O_3$	$73.6934 + 0.031079972 \text{ x T} - (1555796)/T^2$	2.8613
$(CaO)_4Al_2O_3Fe_2O_3$	$72.708592 + 0.039229 \text{ x T} - (1250350{,}86)/T^2$	6.4143
$(CaO)_2SiO_2$	$45.8964 + 0.01968989 \text{ x T} - (735262)/T^2$	1.5805
$(CaO)_3SiO_2$	$44.884 + 0.01982367 \text{ x T} - (665117.42)/T^2$	3.5810

The most common fuels in rotary kilns are charcoal, mineral coal, heavy oils and petroleum coke (or just coke). In Brazil the coke is one of the most widely used in the cement industry. Its composition is shown in Table 2 (Szargut et al., 1988; Wall, 1990).

Table 2. Composition of petroleum coke and its enthalpy

Fuel	**Carbon (C)**	**Hydrogen (H)**	**Oxygen (O)**	**Nitrogen (N)**	**Sulfur (S)**	**ΔH [MJ/kg]**
Petroleum coke	97.5	0.3	0.3	1.0	0.9	33.2

The exergy calculation is computed as follows in Equation 13 to 15 (Koroneos et al, 2003).

$$B_{ch_{coke}} = \Delta H_{coke} \cdot \left(1{,}0437 + 0{,}1896 \cdot \frac{x_H}{x_C} + 0{,}0617 \cdot \frac{x_O}{x_C} + 0{,}0428 \cdot \frac{x_N}{x_C}\right) + 9710 \cdot x_s \quad (13)$$

Where x_C, x_H, x_O, x_N, x_S are the molar fractions of the elements on coke, and B_{ch} is the chemical exergy of the component subscript.

The calculations are made based on kg of fuel with the formula $C_\alpha H_\beta N_\gamma O_\delta S_\varepsilon$, which passes through the combustion reaction presented on Equation 14.

$$C_\alpha H_\beta N_\gamma O_\delta S_\varepsilon + \frac{2\cdot(\alpha+\varepsilon)-\delta+\frac{\beta}{2}}{2} \cdot O_2 \rightarrow \alpha \cdot CO_2 + \frac{\beta}{2} \cdot H_2O_{liq} + \frac{\gamma}{2} \cdot N_2 + \varepsilon \cdot SO_2 \quad (14)$$

Where $\alpha = x_C/12$, $\beta = x_H/1$, $\delta = x_O/16$, $\gamma = x_N/14$, $\varepsilon = x_S/32$.

The Gibbs free energy (ΔG) variation per kg of coke combustion is given by Equation 15, which is the exergy variation of these reactions.

$$\Delta G_{Fuel} = \alpha \cdot B_{ch_{CO_2}} + \frac{\beta}{2} \cdot B_{ch_{H_2O_{liq}}} + \frac{\gamma}{2} \cdot B_{ch_{N_2}} + \varepsilon \cdot B_{ch_{SO_2}} - B_{ch_{Fuel}} - \frac{2\cdot(\alpha+\varepsilon)-\delta+\frac{\beta}{2}}{2} \cdot B_{ch_{O_2}} \quad (15)$$

2.3.2 CO_2 capture

The CO_2 capture process in this study was taken as that using monoethanolamine (MEA) in the process of absorption and desorption. The inputs on the CO_2 capture process are the combustion gases from the cement plant and steam for reboiling the MEA solution (Jassim; Rochelle, 2006). The outputs are the streams of combustion gases poor in CO_2 (8% of the total CO_2 input in this process), the CO_2 captured and separated and the water output from the reboiler. Inputs and outputs of energy from the intermediate processes were neglected for approximation purpose. Figure 2 indicates such control volume.

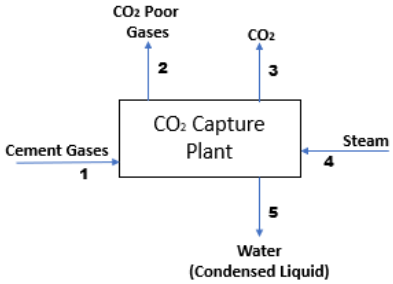

Figure 2: Control Volume of the CO_2 Capture Plant

The composition on each stream can be seen in Table 3.

Table 3. Mass flow in each stream on the CO_2 capture process

Stream	Stream Mass Flow (kg/tonne of clinker)					
	H_2O	O_2	CO_2	Ar	N_2	Total
1	3.296×10^2	2.093×10^2	1.380×10^3	5.263×10^1	3.116×10^3	5087.41
2	3.296×10^2	2.093×10^2	2.071×10^2	5.263×10^1	3.116×10^3	3914.16
3	1.660×10^1	0	1.173×10^3	0	0	1189.85
4	1.760×10^3	0	0	0	0	1759.71
5	1.760×10^3	0	0	0	0	1759.71

2.3.3 Electrolysis

The electrolysis plant breaks water into H_2 and O_2 as represented in Figure 3. The chosen process was PEM electrolysis which is adequate for intermittent energy sources as wind-based. The reactions of PEM water electrolysis are expressed in Equations 16 to 18 (Carmo et. al, 2013).

Cathode: $2\ H^+ + 2\ e^- \rightarrow H_2$ (16)

Anode: $H_2O \rightarrow 2\ H^+ + ½\ O_2 + 2\ e^-$ (17)

Global: $H_2O \rightarrow H_2 + ½\ O_2$ (18)

It was taken for the present study the electrolysis at 1 bar, input temperature of 368 K for the water and output temperature of 353 K for H_2 and O_2 (Ani; Agachi, 2007). The electrical energy consumption on the process was considered equal to 7.5 kWh/m³ H_2 or its equivalent on the plant, of 3.87×10^{13} kJ/year (Carmo et. al, 2013).

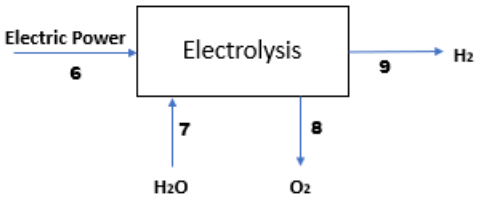

Figure 3: Control Volume of the Electrolysis Plant

The composition on each flow can be seen at Table 4.

Table 4. Flow in each stream on electrolysis process

Element	**Stream**	**Flow**	**Unit**
Electric power	6	3.87×10^{13}	kJ/year
H_2O	7	8.79×10^5	t/year
O_2	8	7.81×10^5	t/year
H_2	9	9.86×10^4	t/year

2.3.4 Methanation

The methanation process turns the CO_2 and H_2 inputs, provenient from the cement industry and electrolysis respectively, into methane, water and heat (because it is an exothermic reaction) as it can be seen in Figure 4. In the present study it was used the pressure of 20 bar and a temperature of 300 ºC, values usually found in literature. The heat associated with the reaction was assumed as 165 kJ/mol of reaction as it can be seen in Equation 19 (Lehner et al, 2014).

$$CO_{2\,(g)} + 4\,H_{2\,(g)} \leftrightarrow CH_{4\,(g)} + 2\,H_2O_{(g)} \qquad -165\ kJ/mol \tag{19}$$

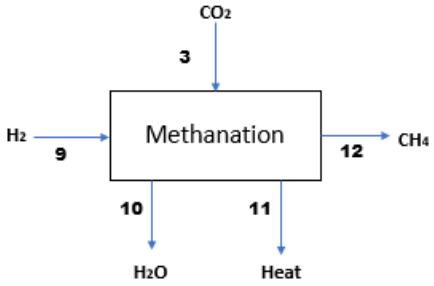

Figure 4: Control Volume of the Methanation Plant

The composition on each stream can be seen at Table 5 and the stream 11 takes the enthalpy of the reaction of Equation 19.

Table 5. Mass flow in each flow on electrolysis process

Element	Stream	Mass Flow (t/year)
CO_2	3	5.73×10^5
H_2	9	9.86×10^4
H_2O	10	4.40×10^5
CH_4	12	1.96×10^5

2.3.5 Intermediary Compressions

The intermediate compressions are used for even the pressures of the outputs from one process that become the inputs of other processes. It is used to pressurize the CO_2 from 1 bar and 298K and the H_2 from 1 bar and 353 K both to 20 bar and 573 K for the input of the methanation process. A more detailed analysis and the efficiency of the compressions was estimated as 85% can be found in literature (Mady et. al, 2018).

2.4 Results and discussions

Using the data from section 2.3 and through the Equations (1) to (4) it was possible to obtain the gross destroyed exergy and the exergy efficiency as follows in Table 6.

Table 6. Destroyed exergy and exergy efficiency per process

Process	B_d (kJ/year)	Exergy Efficiency (%)
Cement – Crushing and milling	3.75×10^{16}	~ 0
Cement Production	1.98×10^{12}	56
CO_2 Capture	7.83×10^{12}	46
Electrolysis	2.84×10^{12}	32
Methanation	1.24×10^{12}	89

The electric power needed for the processes (excluding crushing and milling) was calculated as being 3.62×10^{13} kJ/year. Therefore, the relation between the electric power needed to transform the CO_2 produced on CH_4 is of 67.3 MJ/kg of CO_2.

The results of crushing and milling the materials in the cement industry were considered near 0% because of the conversion of electric energy on physical energy to crush the materials. There is a conversion of high-quality energy into low-quality energy.

On cement production, the efficiency result of 56% is compatible with those on literature, of 50 to 68% (Koroneos et. al, 2003) and 38.68% (Jijesh et. al, 2015). Though it is necessary to notice that the literature methods used other methodologies, data and approximations.

The exergy efficiency of 46% on the CO_2 capture process is 20% percentage difference from the 38.32% found in the literature (Xie et. al, 2017). This difference may be due to the process simulation on literature, which was carried using a different control volume and the software AspenPlus. Moreover, the compression with intercooling may decrease considerably the values of exergy efficiency (increase in the destroyed exergy).

The electrolysis process with operating conditions of 80ºC and 1 atm in this study has an exergy efficiency of 32% and those found in the literature for the same conditions have exergy efficiency of 62,18% (Yilmaz et. al, 2014) and 56.5% (Ahmadi et. al, 2013). As known, the efficiency of an electrolysis system depends on the pressure of the system, its temperature and the density of the current applied. In this regard, the electric current conditions shall be taken in consideration for a better comparison.

The catalytic methanation in this study has an exergy efficiency calculated as 89%. However, there are few studies to be compared to this value due to the recent arise of this technology. This process can be utilized in various processes with different types of inputs, but all with the goal of producing methane. In a process of catalytic methanation using biomass, the exergy efficiency is around 70% and can reach 75.4% (Gassner et. al, 2011).

Regarding to the exergy efficiency of the complete plant, it is almost zero because of the irreversibilities associated with the crushing and milling processes. If these processes are not included, the exergy efficiency of the entire plant is of 48%. This percentage reflects the fact that more than half of the energy input is wasted in internal processes like heat losses and internal irreversibilities through the conversion of energy.

The exergy efficiency conversion and destruction (excluding the cement crushing and milling) is presented in the Grassmann diagram in Figure 5. It shows the data of Table 6, in which the percentage of losses is defined by the stream input on its respective process. This diagram gives a better dimension and understanding of the energy irreversible conversions.

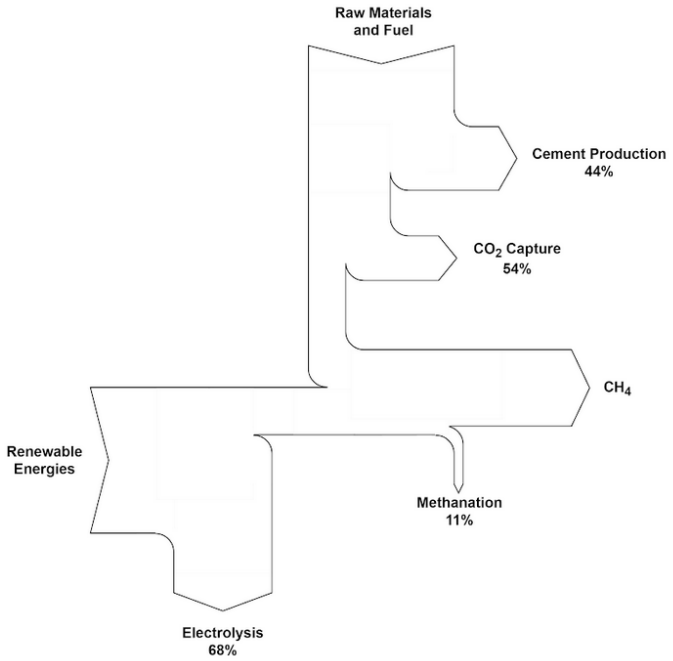

Figure 5: Grassmann Diagram with percentages of exergy destruction and conversion

3. CONCLUSIONS

In this study the goal of generating CH_4 for storage (aiming its reconversion to electrical energy to grid balance when needed) integrated the process of methanation with those of cement production and carbon capture as well the electrolysis and energy generation. These processes were analyzed in its separated plants and combined in a power-to-gas plant with CO_2 input from a cement factory. The operational data of each plant were defined according to literature and the procedures approximated to the real ones through the definition of control volumes that contains the inputs and outputs of the real processes.

The stoichiometric relations between the processes revealed that the energy consumption per kg of CO_2 input on the process is of 67.3 MJ. Also, to the adopted information and data and through the second law of thermodynamics analysis, it was possible to obtain the exergy efficiency of the processes separately and the whole joint plant. The results provide an approximation of the exergy efficiency of 48% for the whole plant (without the crushing processes in cement production), which reveals the irreversible characteristics of the processes. Furthermore, it is possible to notice the process with higher energy degradation, which are the physical crushing of materials for cement production, followed by the electrolysis process.

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