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EXERGY ANALYSIS IN HYDROGEN PRODUCTION FROM AUTOTHERMAL REFORMING OF NATURAL GAS

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Abstract: This work makes an exergetic analysis in the production of hydrogen by Autothermal Reforming of Methane, ATR. ATR is combination between Steam Methane Reforming, SMR, and partial reforming. In the ATR, the reforming reaction can be endothermic, exothermic or neutral depending on the relation CH_4-H_2O-O_2. The first step of ATR is the reforming where synthesis gas, containing H_2, H_2O, CO_2, CO and residual CH_4 is formed. Thermodynamic chemical equilibrium is assumed in all reactions of process. After reforming, Shift reactions produce more H_2 reacting CO and H_2O. After Shift Reactions the synthesis gas is purified by absorption and adsorption. The absorption is made in Diethanolamine, DEA, and produce CO_2 as by product. The adsorption is made in fixed beds of molecular sieves. This procedure can produce H_2 highly pure (99,999 %, wt) proper to fuel cells. The exergetic analysis identifies the sources of irreversibility of the process. It was employed the Fuel-Product concept where the Control Volumes, CV, may present as product the total, physical or chemical exergies. The desegregation of the exergy into physical and chemical is essential to evaluate the perform of CV's. The results show that the main source of irreversibility is the Reformer.

Keywords: *Hydrogen Production, Exergy; Natural Gas, Autothermal Reforming*

1. INTRODUCTION

The increasing availability of natural gas in Brazil opens many possibilities of use in many fields of economy. The main use of hydrogen is in refinery to hydrotreatment of fuels. Its main application in the industrial sector is as fuel to processes or to electric generation. The commercial, domestic and vehicular is still a small parcel but, that has grown and will be significant in next years (CTPETRO 2001).

One promissory use of hydrogen is in fuel cells. Fuel cells have high efficiency in conversion to electric energy. The efficiency is higher than other conventional methods. Although its high efficiency, the systems have low power by volume being necessary larger systems to produce an amount economically viable of energy. With development of fuel cells the use of hydrogen must be intensified, mainly to residences, vehicles and small electric installations (Kothari et al., 2004).

ATR is an advance in relation the SMR, the main difference is that ATR promotes the reaction between CH_4 and H_2O in presence of O_2 inside Reformer tubes. The reaction between O_2 and CH_4 liberates energy to drive the endothermic reaction between H_2O and CH_4. This process as well as SMR can be used to produce highly pure hydrogen, by about 99,999 % (wt) due to purification step in adsorption beds.

This work is part of a bigger one that performs an exergoeconomic analysis of processes of production of hydrogen (Alves, 2007). In this work the simulation of the process of hydrogen production by ATR is described and the exergy determined in each step. The ATR process aims to diminish the irreversibility of the combustion chamber as source of heat to promote the reaction.

2. PROCESS DESCRIPTION

Similar to SMR, ATR comprises three basic stages: the reform, the Shift Reaction and the purification of hydrogen. It was assumed equilibrium hypothesis for chemical reactions, absorption and adsorption. The purge gas, exiting the adsorption step, has a considerable quantity of H_2. The purge gas is conducted to the combustion chamber, where it is burned to save CH_4. Figure 2 shows a schematic of ATR process.

To improve the reforming, O_2 is injected inside reformer. An unit of separation of oxygen, running on standard Skarstrom cycle provides this O_2 to Reforming.

The inlet conditions of methane, water and air are: 4 atm and 25ºC to methane and 1 atm and 25ºC to air and water. Besides the material inputs the process also requires electricity to drive the auxiliary equipment such as pumps and compressors.

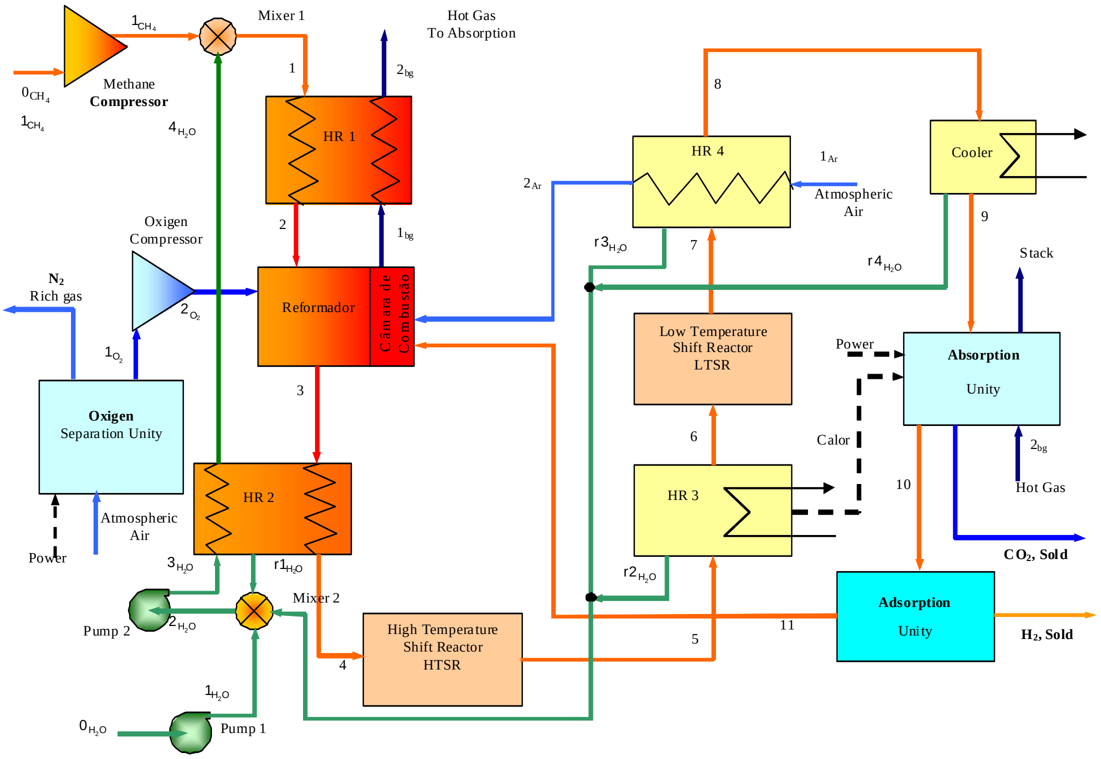

Figure 1 – Schematic of ATR process.

2.1. Autothermal Reforming

After mixed, H_2O, CH_4 and O_2 enter in Reformer, composed by a fixed bed with nickel catalyst.
The autothermal reforming comprises three basic reactions:

$$CH_4 + 2\ O_2 \rightarrow CO_2 + 2\ H_2O \tag{1}$$
$$CH_4 + H_2O \rightarrow CO + 3\ H_2 \tag{2}$$
$$CO + H_2O \rightarrow CO2 + H2 \tag{3}$$

The reaction (1) is exothermic and liberates energy to drive the further reactions (endothermic). These reactions can be globally exothermic, endothermic or neutral, depending on the amount of O_2 injected in reformer and the ratio H_2O-CH_4. In this work, the reactions are globally endothermic. At the end of process, the purge gas can be conducted to combustion chamber of Reformer saving methane.

The Reformer works with two heat exchange, HR1 and HR2. HR1 preheat the gases before enter in reformer, HR2 cool the synthesis gas to enter the Shift Reactor and generate steam to mix with CH_4.

The unit of production of oxygen operates according to Skartrom cycle (PSA). The consumption of power, working temperature and recuperation factor was assumed according to Mendes et al. (2001).

2.2. Shift Reaction

The Shift reactions occur in two successive steps in fixed bed reactor with iron catalyst. Shift reaction is the reaction between the existing CO and H_2O in synthesis gas. This reaction is exothermic and heats the synthesis gas more in the first reactor than in the second. Due to this behavior, the first Shift Reactor is named High Temperature Shift Reactor and the second Low Temperature Shift Reactor.

2.3. Absorption Unit

In this unit the CO_2 is removed from synthesis gas, increasing the concentration of H_2 and generating CO_2 as an important by-product. The absorption is made by a solution of Diethanolamine, DEA.

To improve efficiency in the absorption process the gas must be cooled before absorption column and heated before the desorption column. The coolers are heat exchanges that liberate heat to the atmospheric air. The heaters are heat exchanges that use the energy of process to save fuel in this unit. A schematic of Absorption Unit can be seen in Fig 2.

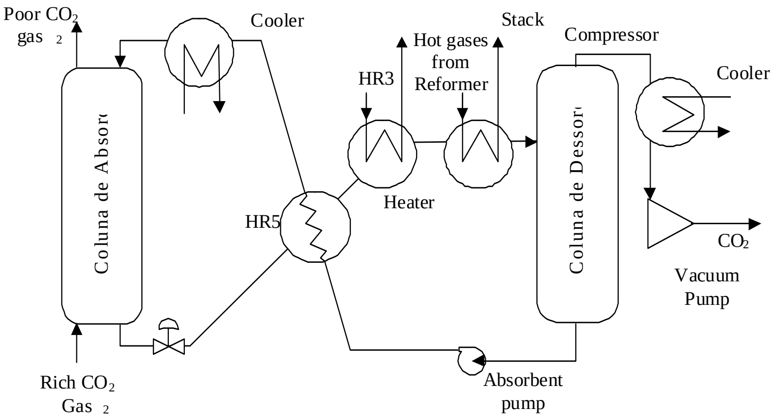

Figure 2 – Schematic of absorption unit operating with Diethanolamine

The temperature of desorption is 40ºC. For this temperature the minimum pressure is 40 kPa. The absorption desorption were simulated according to correlations of Kent & Eisenberg (1976), modified by Park et al (2002).

2.4. Adsorption Unit

Adsorption is the last step of hydrogen production; it purifies the H_2 up to 99,999% (wt). This unit operates in a PSA cycle where a system of valves divides the upstream gas into two streams, the pure H_2 and the purge gas. The Recuperation factor, understood as amount of hydrogen present in the stream that is separated after adsorption cycle, was assumed as 82%. This value was based on Yang (1997) and Sircar et al. (1999). The hydrogen exits adsorption process at 40ºC and 1200 kPa and purge gas at 101 kPa and 40ºC. For this purge gas a Low Heat Value of 25400 kJ/kg was calculated.

3. Exergetic Analysis

In order to improve exergetic analysis, the process was divided into several control volumes CV and each performance relative to its function was determined. Some considerations were assumed:

- Environmental standard temperature and pressure conditions T0=25ºC and P0=101,3 kPa to exergy calculus. The composition was assumed as that proposed by Szargut (1988).
- Efficiencies determined according to Fuel F, Product P and Irreversibility I concept of Kotas (1985), ε, or input output method, ζ, for dissipative control volumes. These efficiencies are defined as:

Methane Compressor, being $\dot{W}_{CH_4,Comp}$ the power required by the compressor:

$$F = \dot{W}_{CH_4,Comp} \tag{4}$$

$$P = Ex_{1,CH_4} - Ex_{0,CH_4} \tag{5}$$

Water pump 1:

$$F = \dot{W}_{pump,1} \tag{6}$$

$$P = Ex_{1,H_2O} - Ex_{0,H_2O} \tag{7}$$

Water pump 2:

$$F = \dot{W}_{pump,2} \tag{8}$$

$$P = Ex_{3,H_2O} - Ex_{2,H_2O} \tag{9}$$

Being $\dot{W}_{pump,1}$ e $\dot{W}_{pump,2}$ the power of pump 1 and 2.

Heat Recovery 1 (HR1):

$$F = Ex_{1,bg} - Ex_{2,bg} \tag{10}$$
$$P = Ex_2 - Ex_1 \tag{11}$$

Inside HR2 condensed water can be exist. In this case, there is a change of chemical exergy. Equation (12) gives the global formulation for the Fuel in HR2 and equations (14) and (15) disaggregates into physical and chemical. The HR2 balance is:

$$F = Ex_3 - Ex_4 - Ex_{r1,H_2O} \tag{12}$$
$$P = Ex_{4,H_2O} - Ex_{3,H_2O} \tag{13}$$
$$F_{Ph} = Ex_{Ph3} - Ex_{Ph4} - Ex_{Phr1,H_2O} \tag{14}$$
$$F_{Ch} = Ex_{Ch3} - Ex_{Ch4} - Ex_{Chr1,H_2O} \tag{15}$$

Heat Recovery 4 (HR4):

$$F = Ex_7 - Ex_8 - Ex_{r3,H_2O} \tag{16}$$
$$P = Ex_{2;Ar} - Ex_{1;Ar} \tag{17}$$

To reformer it is possible define one chemical fuel and two products, one chemical and one physical:

$$F = (Ex_{11} + Ex_{2,Ar}) - Ex_{1,bg} \tag{18}$$
$$P = Ex_3 - Ex_2 - Ex_{2,O_2} \tag{19}$$
$$F_{Ch} = (Ex_{Ch,11} + Ex_{Ch1,Ar}) - Ex_{Ch1,bg} \tag{20}$$
$$P_{Ch} = Ex_{Ch3} - Ex_{Ch2} - Ex_{Ch2,O_2} \tag{21}$$
$$P_{Ph} = Ex_{Ph3} - Ex_{Ph2} - Ex_{Ph2,O_2} + Ex_{Ph1,bg} - Ex_{Ph2,Ar} - Ex_{Ph11} \tag{22}$$

The balance of Unit of Oxygen Separation follows, being $\dot{W}_{O_2,Sep}$ the power required to oxygen separation:

$$F_{O_2,Sep} = \dot{W}_{O_2,Sep} \tag{23}$$
$$P_{O_2,Sep} = Ex_{1,O_2} \tag{24}$$
$$P_{Ch,O_2,Sep} = Ex_{Ch1,O_2} \tag{25}$$
$$P_{Ph,O_2,Sep} = Ex_{Ph1,O_2} \tag{26}$$

The balance of Oxygen Compressor, being $\dot{W}_{O_2,Comp}$ the Power required by the O_2 compressor:

$$F_{O_2,Sep} = \dot{W}_{O_2,Comp} \tag{27}$$
$$P_{O_2,Sep} = Ex_{Ph2,O_2} - Ex_{Ph1,O_2} \tag{28}$$

High Temperature Shift Reactor (HTSR):

$$F = Ex_{ch,4} - Ex_{ch,5} \tag{29}$$
$$P = Ex_{ph,5} - Ex_{ph,4} \tag{30}$$

Low Temperature Shift Reactor (LTSR):

$$F = Ex_{ch,6} - Ex_{ch,7} \tag{31}$$
$$P = Ex_{ph,7} - Ex_{ph,6} \tag{32}$$

The function of mixers is to homogenize the gas. Mixers are dissipative CVs, they destroy chemical and physical exergy. To mixer 1 the balance is:

$$Ex_e = Ex_{4,H_2O} + Ex_{1,CH_4} \quad (33)$$

$$Ex_s = Ex_1 \quad (34)$$

Mixer 2:

$$Ex_e = Ex_{r1,H_2O} + Ex_{r2,H_2O} + Ex_{r3,H_2O} + Ex_{r4,H_2O} + Ex_{1,H_2O} \quad (35)$$

$$Ex_s = Ex_{2,H_2O} \quad (36)$$

Assuming Absorption Unit like a dissipative CV, including the cooler and HR3, it is possible to write the exergy flows that enter and exit.

$$Ex_{e,Abs} = (Ex_5 - Ex_6) + Ex_8 + Ex_{2,bg} + \dot{W}_{Bomba,ABS} + \dot{W}_{Comp,CO_2} \quad (37)$$

$$Ex_{s,Abs} = Ex_{10} + Ex_{CO_2,Sold} + Ex_{r2,H_2O} + Ex_{r4,H_2O} + Ex_{3,bg} \quad (38)$$

Being $\dot{W}_{Bomba,ABS}$ e $\dot{W}_{Comp,CO_2}$ the required power of absorbent pump and CO_2 Compressor, respectively.

Defining the product of this unit as the input of chemical exergy in the gases that exit, CO_2 and process gas, it is possible to write for this unit its fuels and products with the decomposed exergy:

$$F_{\dot{W}} = \dot{W}_{ABS,Pump} + \dot{W}_{CO_2,Comp} \quad (39)$$

$$F_{Ph} = (Ex_{ph5} - Ex_{ph6}) + (Ex_{ph2,bg} - Ex_{ph3,bg}) + Ex_{Ph8} - (Ex_{Ph10} + Ex_{CO_2} + Ex_{phr2,H_2O} + Ex_{phr4,H_2O}) \quad (40)$$

$$F_{Ch} = (Ex_{Ch5} - Ex_{Ch6}) \quad (41)$$

$$P_{Ch} = Ex_{Ch10} + Ex_{CO_2} - Ex_{Ch8} + (Ex_{Chr2,H_2O} + Ex_{Chr4,H_2O}) \quad (42)$$

In heat exchanger HR3 there is condensed, therefore the chemical exergy Ex_{Ch5} and Ex_{Ch6} are different and its variation configures as chemical fuel for the unit of absorption

Adsorption Unit:

Assuming this unit as a dissipative CV it is possible to write the exergy flows that enter and exit:

$$Ex_e = Ex_{10} \quad (43)$$

$$Ex_s = Ex_{11} + Ex_{H_2} \quad (44)$$

Assuming the product of this unit as the input of chemical exergy in the gases that exit, H_2 and purge gas, it is possible to write for this unit its fuels and products with the decomposed exergy:

$$F = Ex_{Ph10} - (Ex_{Ph,H2} + Ex_{Ph,11}) \quad (45)$$

$$P = (Ex_{Ch,H2} + Ex_{Ch,11}) - Ex_{Ch,10} \quad (46)$$

Global Process:

In the process two forms of exergy enter, material flows and electric power. These two forms are discriminated as below power exergy, $Ex_{e,\dot{W}}$, and exergy due to the material flow, $Ex_{e,m}$, and the exergy that exits the process, Ex_s:

$$Ex_{e,\dot{W}} = \dot{W}_{CH_4,Comp} + \dot{W}_{Pump,1} + \dot{W}_{Pump,2} + \dot{W}_{O_2,Sep} + \dot{W}_{O_2,Comp} + \dot{W}_{Pump,ABS} + \dot{W}_{CO_2,Comp} \quad (47)$$

$$Ex_{e,m} = Ex_{0,H_2O} + Ex_{0,Ar} + Ex_{1,CH_4} \quad (48)$$

$$Ex_s = Ex_{CO_2} + Ex_{H_2} \quad (49)$$

The temperature, pressure and mass flow along of process as well as the exergies had been calculated related to 0,01 kmol/s of methane that enters in the reformer. Table 1 shows these values.

Table 1 – Operational Parameters of ATR, (0,01 kmol/s of CH_4 entering in Reformer)

Flow	T (K)	P (kPa)	$\dot{N}$ (kmol/s)	Flow	T (K)	P (kPa)	$\dot{N}$ (kmol/s)
0_{H_2O}	298,2	101,3	0,01343	0air (O2 Sep)	298,2	101,3	0,01515
1_{H_2O}	298,3	1926	0,01343	1_{O_2}	298,2	101,3	0,003182
2_{H_2O}	373,2	1887	0,04	2_{O_2}	802,1	2856	0,003182
3_{H_2O}	373,4	3150	0,04	1	1045	2885	0,05
4_{H_2O}	1203	3000	0,04	2	1104	2596	0,05
$r1_{H_2O}$	418,4	2109	0,02331	3	1223	2337	0,06978
$r2_{H_2O}$	370	1984	0,002329	4	417,3	2220	0,04647
$r3_{H_2O}$	366,8	1867	0	5	538,3	2153	0,04647
$r4_{H_2O}$	313,2	1830	0,000931	6	370	2089	0,04414
0_{CH_4}	298,2	405,3	0,01	7	394,8	2026	0,04414
1_{CH_4}	480,2	3000	0,01	8	367,3	1965	0,04414
1bg	1273	101,3	0,02069	9	303	1926	0,04321
2bg	1085	101,3	0,02069	10	303	1733	0,03372
3bg	412,2	101,3	0,02069	11	303	101,3	0,006625
0ar (cc)	298,2	101,3	0,01712	CO2	458,7	101,3	0,00949
1air	3734,8	101,3	0,01712	H2	303,2	1416	0,0271

Table 2 shows the molar composition of the main flows between the reformer and the exit of the adsorption unit. The exergetic efficiencies of CV are shown in Table 3; the required power in Table 4 and; the exergies, total, physical and chemical in Table 5.

Table 2 – Molar Composition of main flows

Fluxo	CH_4	CO	CO_2	H_2	H_2O	O_2	N2
1	0,2	–	–	–	0,8	–	–
3	0,001553	0,07877	0,06298	0,397	0,4597	–	–
4	0,002333	0,1183	0,09457	0,5962	0,1886	–	–
5	0,002333	0,02094	0,1919	0,6935	0,09131	–	–
6	0,002456	0,02204	0,202	0,7301	0,04338	–	–
7	0,002456	0,003485	0,2206	0,7486	0,02482	–	–
8	0,002456	0,003485	0,2206	0,7486	0,02482	–	–
9	0,002509	0,00356	0,2253	0,7648	0,003808	–	–
10	0,003215	0,004562	0,007979	0,98	0,004232	–	–
11	0,01637	0,02323	0,04063	0,8982	0,02155	–	–
Bg	–	–	0,01798	–	0,3049	0,6536	0,0158

According to expected, the biggest source of irreversibility is the reformer, 46% of net irreversibility of the process. This characteristic is due to the combustion that naturally generates a lot of irreversibility, as can be seen in Alves and Nebra (2003).

Another great source of irreversibility is the absorption unit. Although the gases that goes to stack has no utility, 3bg, it does not contribute to irreversibility balance of the unit. The high irreversibility of this unit is due to heat and electric energy required. The irreversibility of stream 3_{bg} does not penalize adsorption unit but the stack so is part of net irreversibility of process.

The definition of exergetic product used in absorption and adsorption units resulted in low exergetic efficiencies although if the methodology to dissipative CVs is adopted these efficiencies are high.

Tabela 3 – Exergia dos fluxos, com referência a 1 kmol/s de metano que entra no reformador.

Flow	Ex_{ph} (kW)	Ex_{ch} (kW)	Ex (kW)	Flow	Ex_{ph} (kW)	Ex_{ch} (kW)	Ex (kW)
0_{H_2O}	0	12,09	12,09	1ar	4,124	0	4,124
1_{H_2O}	0,4185	12,09	12,51	1_{O_2}	0	12,63	12,63
2_{H_2O}	25,73	36	61,73	2_{O_2}	47,65	12,63	60,28
3_{H_2O}	26,81	36	62,81	1	1472	8290	9763
4_{H_2O}	1442	36	1478	2	1578	8290	9869
$r1_{H_2O}$	34,96	20,98	55,94	3	1805	8344	10149
$r2_{H_2O}$	1,393	2,096	3,488	4	383,3	8186	8569
$r3_{H_2O}$	0	0	0	5	448,9	8083	8532
$r4_{H_2O}$	0,05411	0,838	0,8922	6	341,3	8076	8418
0_{CH_4}	34,37	8317	8351	7	345,3	8065	8410
1_{CH_4}	100,6	8317	8417	8	333,8	8065	8399
1bg	391	40,44	431,5	9	315,7	8066	8382
2bg	267,2	40,44	307,6	10	237,6	7931	8169
3bg	3,958	40,44	44,4	11	0,06959	1536	1536
0ar (cc)	0	0	0	CO_2	14,09	188,1	202,2
				H_2	167	6398	6565

Tabela 4 – Consumo de potência de acionamento pelas unidades

Volume de Controle	$\dot{W}$ (kW)	$\dot{W}$ (%)
CH_4 Compressor	73,2	27,6
O_2 Compressor	50,61	19,1
O_2 Unit	56,98	21,5
Water Pump 1	0,5231	0,2
Water Pump 2	1,277	0,5
Absorbent Pump	22,03	8,3
CO_2 Compressor	60,42	22,8
Total	265,0	100

Tabela 5 – Eficiência exergética dos volumes de controle

Volume de Controle	Ir (kW)	Ir (% Total)	ε	ζ
HR1	17,91	0,9625	0,8554	
HR2	108,2	5,816	0,929	
HR4	7,385	0,3969	0,3583	
HTSR	37,52	2,017	0,6363	
LTSR	7,228	0,3885	0,3577	
Mixer 1	132,6	7,128		0,9866
Mixer 2	11,09	0,5961		0,8477
O_2 Compressor	2,956	0,1589	0,9416	
O_2 Unit	44,35	2,383	0,2217	
CH_4 Compressor	6,998	0,3761	0,9044	
Water Pump 1	0,1046	0,005621	0,8001	
Water Pump 2	0,2039	0,01096	0,8403	
Reformer	889	47,78	0,1982	
Absorption Unit	482,9	25,95	0,1064	0,9721
Adsorption Unit	67,76	3,642	0,03956	0,9917
Stack	44,4	2,386		
Global	1861	100		0,778

In the adsorption unit the increase of chemical exergy was small related to heat and power required by this unit. This system has a low exergetic efficiency by the Fuel Product method.

The net process efficiency was 77,8%, slightly higher than SMR, 77% (Alves, 2007).

The required power in ATR process is to drive the compressors. The methane compressor requires 27,6% of total consumption, the second biggest consumption is to CO compressor, 22,8%.

4. CONCLUSIONS

The operation of Reformer is endothermic to profit of calorific power of purge gas that can be used in combustion chamber saving methane and pure oxygen.

The hot gases that exit combustion chamber have not enough conditions to heat the absorbent of the absorption unit, being been necessary to dislocate exchanger HR3 for inside of this unit.

The system of recycle condensed water along the process was a decisive factor to maintain the sated ration of steam to methane with minor capitation of new water.

The net process efficiency was of 77,8%, superior to SMR, 75.6% (Alves, 2007).

The Fuel Product efficiency used for the units of absorption and adsorption resulted in very efficiencies in these CVs. When applied the balance between the inlet and outlet exergies, the efficiency is higher.

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