Research Article

Assessment of Energy Performances of a Sulfuric Acid Production Unit using Exergy Analysis

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Abstract

This study deals with the exergetic assessment of a sulfuric acid production unit. It aims to evaluate the performance of the system and show the sources of irreversibilities. The sulfuric acid production unit produces sulfuric acid from liquid sulfur, using double absorption contract process. Such technique is applied to raise the conversion of sulfur dioxide (SO_2) and therefore reduce the emission of SO_2 in the gas rejected to the atmosphere. The results of this study show that the overall efficiency of the studied unit is about 56%. Furthermore a comparison between rational and utilizable efficiencies of the main equipments of the unit is made for the best evaluation of causes of exergy destruction.

Keywords: Sulfuric acid, Exergy, Rational efficiency, Utilizable efficiency. ; Irreversibility

1. Introduction

The rise in the consumption of sulfuric acid and the progressively use of sulfuric acid with higher sulfur concentrations as a raw material basis of the industry, introduced improvements in the contact process technology. The double absorption contact process or also known as the double-catalysis process, patented in 1960 by Bayer, is a significant example of this further development. Higher SO2 conversions and also a reduction of the SO2 emissions are fulfilled (Ullmann's, 2007).

The sulfuric acid process is highly exothermic and the heat recovery is a substantial part of the process. Hence it is important to find practical solutions to evaluate and optimize the energy performances in this process type.

One of the most powerful and important tool of process integration is exergy analysis. This method provides information on the maximum savings that can be achieved by making use of new technologies and new processes. As a complement to material and energy balances, exergy analysis provides a more general and deeper insight into the process, as well as new, unforeseen ideas for improvements (M. Gong , 2005; M. Taklaa et al ,2012).

A number of exergy analyses has been presented including sulfuric acid unit. (D.A. Rasheva and L.G. Atanasova, 2002) made an exergetic study of a double absorption contact process with two steps of conversion and an intermediate absorption with oleum . Moreover, (B.X. Almirall, 2009) has been interested on the exergy analysis of a wet sulfuric acid plant.

The main purpose of this study is to evaluate the energetic performances of a sulfuric acid production unit using a double absorption contact process with four conversion steps and an intermediate absorption with sulfuric acid.

2. Theoretical background and formulation

2.1 Energy analysis

The energy balance for a general steady-flow process assuming steady state and neglecting the contribution from kinetic and potential energy can be written as:

$$0 = Q + W + \sum_i F_i H_i - \sum_e F_e H_e \tag{1}$$

Q and W are respectively the heat and work added to the process and H and F are respectively the specific enthalpy of the material stream and the molar flow rate. Subscripts i and e refer to inlet and outlet streams, respectively (S.C. Kaushika et al, 2011).

2.2 Exergy analysis

2.2.1 Exergy concept

From the thermodynamic point of view, exergy is defined as the maximum amount of work which can be produced by a system or a flow of matter or energy as it comes to equilibrium with a reference environment (E. ESCIUBBA and G. WALL, 2007). Exergy can be consumed or destroyed, due to irreversibility in any real process. Thus exergy analysis is a useful tool for addressing the environmental impact of energy resource utilization, and for furthering the goal of more efficient energy-resource use. For it enables the locations, types and true magnitudes

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of losses to be determined. Also, Exergy analysis reveals whether or not and by how much it is possible to design more efficient energy systems by reducing inefficiencies (M. Gong , 2005).

2.2.2 Exergy balance equation

The basis of the first law of thermodynamics is concept of conservation of energy. Whereas exergy balance equation speaks about degradation of the quality of the energy. These concepts can be clearly demonstrated as below (M. Taklaa et al , 2012); S. M. Sanaei et al, 2012):

Exergy input-Exergy output-Exergy destroyed
-Exergy loss=Exergy accumulation   (2)

Some assumptions are considered, which are: (i) mass flow is one-dimensional, (ii) input and output terms of equation, (2) are net quantities and the accumulation terms are zero. Equation (2) is simplified as (A.R SAIDUR et al, 2007; M Hasanuzzaman et al, 2011):

$$0 = \sum_k Q_k (1 - \frac{T_0}{T_k}) + \sum_l W_l + \sum_i Ex_i - \sum_e Ex_e + Ex_D$$
(3)

The first term on the right hand side is exergy accompanying transfer of thermal energy. The second term can be interpreted as exergy accompanying work and Ex is the exergy rate of the stream of material. Further, Ex_D is the exergy destruction which is directly related to entropy generation with in the process. The created entropy is the difference between the actual process change of entropy and that of its corresponding ideal process. The process inefficiency (irreversibility) is measured as a lost work potential ($Ex_D = T_0 \Delta S$) (S.C. Kaushika et al, 2011; Z. Utluaand A. Hepbaslib, 2007).

The exergy of a stream of material (neglecting contributions from kinetic and potential energy) can be written as the sum of the physical and chemical exergy.
The physical exergy is the maximum work obtainable as the system passes from its initial temperature and pressure, T and P, to the temperature and pressure of the reference environment, T_0 and P_0. At T_0 and P_0, the system is said to be in the restricted dead state. It is given by (N.A. Madloola et al, 2012; A.V Ensinas et al, 2008):

$$Ex_{ph} = F((H - H_0) - T_0(S - S_0))$$
(4)

Chemical exergy (Ex_{ch}) is the maximum work obtainable upon going from the restricted dead state and reaching total equilibrium with the environment. For a real solution, the chemical exergy is given by the following relation (A. Martinez and J. Uche, (2010); J.Szargut et al, 1998)

$$Ex_{ch} = \sum_{j=1}^{m} n_j \left[\varepsilon_0 + RT_0 \ln(x_j \gamma_j) \right]$$
(5)

For gases, it is given by:

$$Ex_{ch} = \sum_{j=1}^{m} n_j \left[\varepsilon_0 + RT_0 \ln(x_j \Phi_j) \right]$$
(6)

γ, Φ and ε denote respectively the activity and the fugacity coefficients and the standard chemical exergy of the components .

2.3 Exergy efficiency

Numerous ways of formulating exergetic efficiency for various energy systems are given in detail elsewhere. It is very useful to define efficiencies based on exergy (sometimes called Second Law efficiencies). Two different approaches are generally used that are (G. WALL, 2007):

- A simple definition of efficiency called 'rational efficiency' is defined as the ratio of the sum of all output exergy terms to the sum of all input exergy terms, So the exergy efficiency $\eta_{ex,r}$ becomes.

$$\eta_{Ex,r} = \frac{\sum Ex_e}{\sum Ex_i}$$
(7)

- Sometimes a part of the exergy going through the system is unaffected. This part of the exergy has been named the transit exergy; Ex_{tr}. A new exergy efficiency called 'Utilizable exergy efficiency ($\eta_{Ex,u}$)for any system is defined as the ratio of the exergy associated with the desired energy output to the exergy associated with the energy expended to achieve the desired output.

$$\eta_{Ex,u} = \frac{\sum Ex_e - \sum Ex_{tr}}{\sum Ex_i - \sum Ex_{tr}}$$

(8)

In this equation Ex_{tr} denotes the transiting exergy which is the part of the exergy entering a unit operation which traverses it without undergoing any transformation and, therefore, is not consumed by that operation (M. Sorin, et al, (1998), J. Szargut, 1993).

3. Process description

The case study process (Figure 1) consists on a sulfuric acid production plant using the double absorption contact process. The process could be divided into four main sections: combustion, conversion, absorption and cooling.
At the first section, the atmospheric air is dried in a drying tower then it is send to the oven. In this device the liquid sulfur is burned with the dried air to produce hot SO2 gases (at 1040 °C), which then are cooled to 423 °C in a waste-heat boiler producing a steam at 60.10^5 Pa. This steam is, then, superheated with the hot gases leaving the first converter bed to produce the high pressure steam.
At the second stage, the cooled SO2 enters into a 4-bed catalytic converter to produce SO3 according to the following reaction.
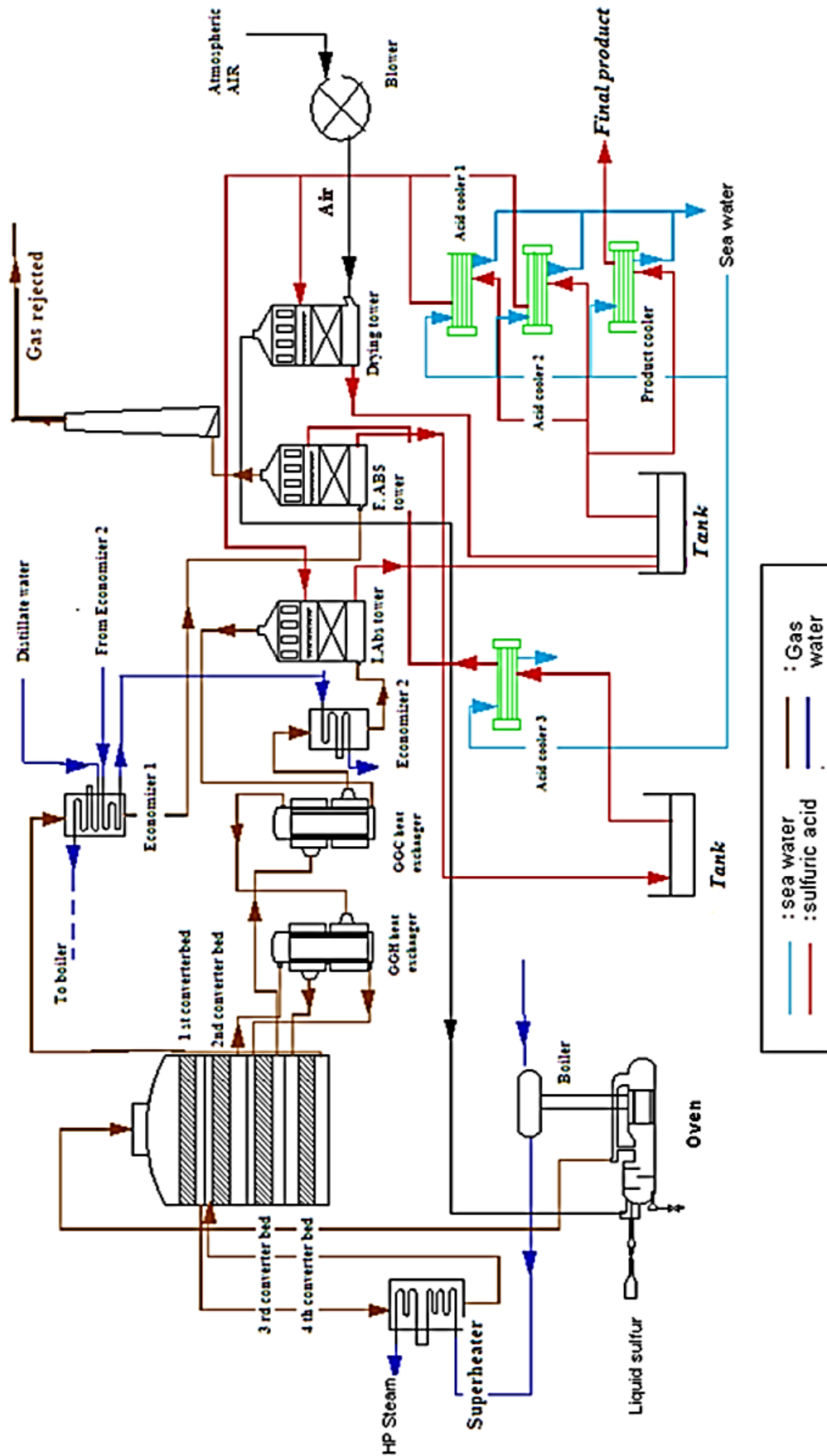
Figure 1: Sulfuric acid production unit.

Table 1: Parameters adopted for the gases streams.

Stream	Temperature (°C)	Pressure (bar)	Molar flow (kmol/h)	Molar Fraction (%)				
				O2	N2	SO2	SO3	H20
Air	40	1,01	6083,64	20,5	77,3			2,2
SO2 from boiler	1040	1,26	5,937,976	9,8	79,2	10,6	0,4	
1st bed converter inlet	423	1,26	5,937,976	9,8	79,2	10,6	0,4	
1st bed converter outlet	607	1,24	5748,51	6,8	81,8	4,4	7	
2nd bed converter inlet	444	1,24	5748,51	6,8	81,8	4,4	7	
2nd bed converter outlet	523	1,21	5,666,409	5,4	83	1,6	10	
3rd bed converter inlet	434	1,21	5,666,409	5,4	83	1,6	10	
3rd bed converter outlet	454	1	5,635,905	4,9	83,5	0,5	11,1	
Economizer 2 inlet	277,4	1	5,635,905	4,9	83,5	0,5	11,1	
Economizer 2outlet	170	1	5,635,905	4,9	83,5	0,5	11,1	
I.Abs tower outlet	83	1	5,008,114	5,5	93,9	0,5		
GGC outlet	316	1	5,008,114	5,5	93,9	0,5		
4th bed converter inlet	433,1	1	5,008,114	5,5	93,9	0,5		
4th bed converter outlet	447	1,07	4,995,506	5,3	94,1		0,5	
Economizer 1outlet	173	1,07	4,995,506	5,3	94,1		0,5	
Gas rejected	77	1,07	4,970,289	5,3	94,6			

Table 2: Parameters adopted for the water streams.

| Stream	Temperature (°C)	Pressure (bar)	Molar flow rate (kmol/h)	Vapor fraction
Economizer 1 inlet	120	66	4641.44	0
Boiler inlet	281.8	66	4641.44	0
Boiler outlet	281.8	66	4636.8	1
Superheated steam	405.4	66	4636.8	1
HP steam	30	3	196030.8	0
Product cooler inlet	30	3	14444	0
Product cooler outlet	34.7	3	14444	0
Cooler 2 inlet	30	3	77777.64	0
Cooler 2 outlet	38	3	77777.64	0
Cooler 1 inlet	30	3	77777.64	0
Cooler 1outlet	38	3	77777.64	0
Cooler 3 outlet	30	3	26031.52	0
Cooler 3outlet	38	3	26031.52	0

$$SO_2(g) + \frac{1}{2} O_2 (g) \rightarrow SO_3(g)$$
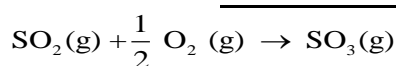
The reaction is highly exothermic and generally it is carried out under adiabatic conditions, so the temperature of the solid catalyst bed rises. The SO2-SO3 equilibrium becomes increasingly unfavorable for SO3 formation as temperature increases (B.X Almirall, (2009), consequently, to achieve a high final SO2 conversion, the total catalyst mass is divided up into four catalyst beds.

The hot gas leaving each bed is cooled, respectively in the superheater, the GGH heat exchanger, the GGC heat exchanger and the economizers, to the minimum working temperature of the catalyst before it enters the next bed.

At the third section, sulfur trioxide gasses are absorbed in both intermediate and final absorption towers. In the intermediate absorption tower (I. Abs tower) gases pass from third bed of conversion, after being cooled, from the

bottom to the top through the absorber, which is uniformly irrigated from the top with liquid-phase sulfuric acid. The H2SO4 liquid stream absorbs the SO3 gasses and it reacts with existing water to form more sulfuric acid with a desired concentration according to the following reaction:

$$SO_3 + H_2O \rightarrow H_2SO_4$$
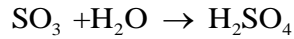
Similarly, in the final absorption tower (F. Abs tower), the gases leaving the forth bed conversion passes from the

bottom to the top to be absorbed by sulfuric acid. The final section is acid cooling where concentrated acid and final product are cooled using see water in acid cooler heat exchangers.

The operational parameters of the main streams of studied unit are represented in tables 1, 2 and 3. In these three tables we present the temperature, pressure, molar flow rate and composition of gases, acid and water streams.

Table 3: Parameters adopted for the acid streams.

Flux	Temperature (°C)	Pressure (bar)	Molar flow rate (kmol/h)	Molar Fraction (%)	
				H_2SO_4	H_2O
Cooler 2 inlet	98	4	12,700,519	93,9	6,1
Cooler 1 inlet	98	4	12,700,519	93,9	6,1
Product cooler inlet	98	4	690,516	93,9	6,1
Produced acid	40	4	690,516	93,9	6,1
Cooler 2 outlet	74	4	12,700,519	93,9	6,1
Cooler 1 outlet	74	4	12,700,519	93,9	6,1
Drying tower inlet	74	3	12,953,583	93,8	6,2
I.Abs tower inlet	82	3	18,738,085	93,9	6,1
I.Abs tower outlet	114	1,01	18,740,194	97,2	2,8
Drying tower outlet	75	1,42	13,087,423	92,9	7,1
Acid cooler 3 inlet	86	4	12,916,113	92,1	7,9
F.Abs tower inlet	77	4	12,916,113	92,1	7,9
F.Abs tower outlet	86	1,01	12,916,113	92,3	7,7

Table 4 :Exergy balance of the sulfuric acid production unit.

Equipments	Exergy in (kW)	Exergy out (kW)	Exergy loss (kW)	% of total losses
Blower	2042,43	1840,45	201,98	0,26
Drying tower	551919,02	551810,22	108,80	0,14
Oven	126229,99	89340,30	36889,69	46,91
Boiler	98217,00	79807,42	18409,58	23,41
Superheater	78570,98	76639,32	1931,66	2,46
Converter	186012,49	184042,91	1969,58	2,50
GGH heat exchanger	64056,14	63734,63	321,51	0,41
GGC heat exchanger	56570,40	54894,63	1675,76	2,13
Economizer 2	52059,64	51785,40	274,24	0,35
Economizer 1	17342,00	16155,81	1186,19	1,51
I.Abs tower	842750,67	832985,57	9765,10	12,42
F.Abs tower	541621,11	540656,68	964,43	1,23
Coolers	1699112,08	1696565,74	2546,34	3.57
Total	4316503,96	4240259,10	76244,86	

4. Exergy analysis of the sulfuric acid unit

4.1 Exergy balance

The exergy analysis of the sulfuric acid production unit was performed according to the following steps:

- Determination of enthalpies and entropies of streams under their actual conditions and those of reference (T_0 =298.15 K et P_0 =1,013 10^5 Pa).

- Calculation of physical and chemical exergy streams using equation 3 and 4 respectively

- Display the results for the total exergy for each stream

$$Ex_{total} = Ex_{ch} + Ex_{ph}$$

- Determination of exergy losses in the equipments using equation 3

- Estimation of exergy efficiencies

The results obtained from the appliance of these steps to the main devices of the studied unit are presented in table 4. From these results we can calculate the contribution of the four sections in the total exergy destruction which are summarized in Figure 2.
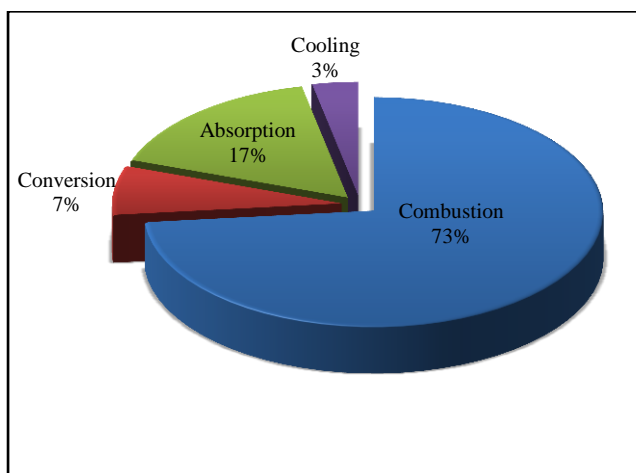

Figure 2: Exergy losses per section.

The exergetic analysis has show that the total exergy losses of the sulfuric acid production unit are around 76 MW with an overall efficiency of 56 %. The sulfur combustion section is responsible for 73 % of the irreversibility generated in the studied unit. The absorption section contributes with 17% of the total losses.

Furthermore 7% and 3% are respectively the parts of the conversion and the acid cooler sections.

The rate of exergy destruction in the oven is about 36 MW thus it is responsible for 47% of the total losses of the unit. These results correspond to an efficiency of about 70%. This can be attributed to the high exothermic reaction.

This irreversible phenomenon leads the augment of temperature between the reactant and product. The temperature difference is the driving force of heat transfer thus the exergy losses are as greater as the temperature difference is greater.

Therefore the recovery boiler contributes with about 23%. This value is a result of the important temperature difference between the feed water and combustion gases. This difference attains 760 ° C.

On the other hand the good thermal insulation of the convertor bed leads to a small contribution of this device in the total loss (about 2.5%).

It should be noted that the absorption of SO2 by sulfuric acid in the towers is irreversible and exothermic. Thus this irreversibility results on about 15% of the total exergy destruction in this step.

4.2 Exergy efficiencies of the main equipments

For the best evaluation of the irreversibility generated in the studied unit we propose to estimate the rational and utilizable efficiencies of the main devices of this unit. An example of calculation of the two efficiencies types are given for the heat exchanger (GGC). A simplified diagram of the exergy balance of this device is given in Figure 3. In this figure $Ex_{C,i}$ and $Ex_{H,i}$ denote respectively the exergy of the cold and hot inlet streams, $Ex_{C,e}$ and $Ex_{H,e}$ design the exergy of the cold and hot outlet streams and Ex_{loss} indicate the exergy loss in the heat exchanger.
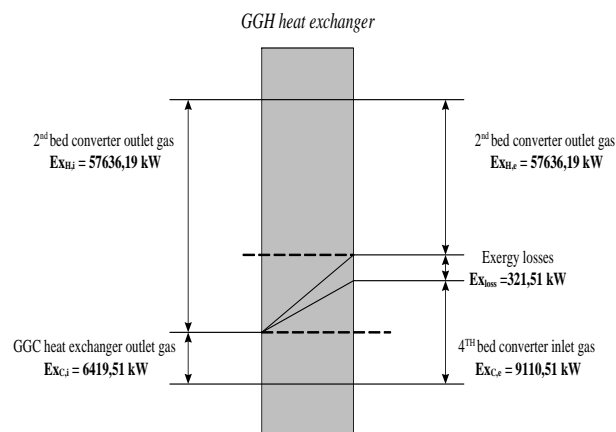

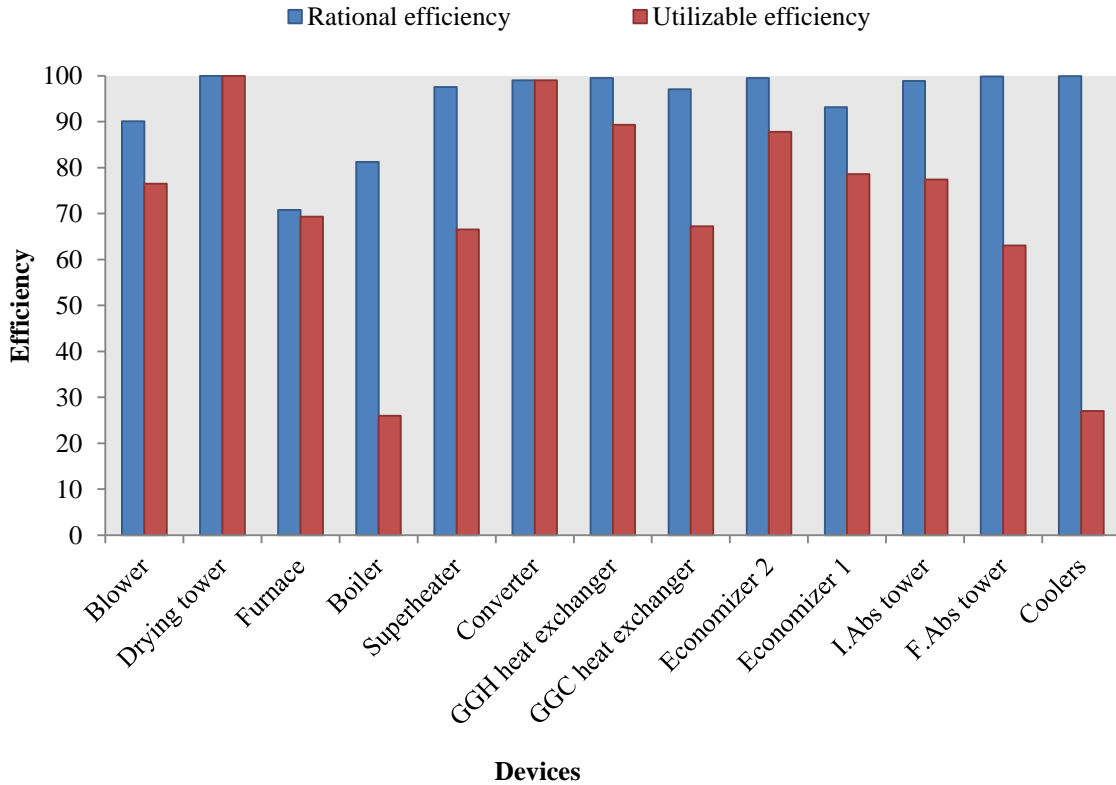Figure 3: Exergy balance diagram of the GGH heat exchanger.

Figure.5: Rational and utilizable efficiencies of the main equipments

The rational efficiency of this exchanger is given by the following relation:

$$\eta_{Ex,r} = \frac{Ex_{H,e} + Ex_{C,e}}{Ex_{H,i} + Ex_{C,i}} \qquad (8)$$

Thus the value of the rational efficiency is about 99%. In the other hand we note from Figure 3 that the quantities $Ex_{C,in}$ and $Ex_{H,out}$ pass through the heat exchanger without changing . These quantities are considered as transit exergy. Therefore the utilizable efficiency is :

$$\eta_{Ex,u} = \frac{(Ex_{H,e} + Ex_{C,e}) - (Ex_{C,i} + Ex_{H,e})}{(Ex_{H,i} + Ex_{C,i}) - (Ex_{C,i} + Ex_{H,e})} \qquad (9)$$

Hence the utilizable efficiency is about 89.33 %.Similarly we calculate the exergy efficiencies of the rest of devices of the sulfuric acid production unit. The results are given in Table5 and the graphical illustration of these results is given by Figure.4.

It arises from these results that utilizable efficiency of the coolers is low even though its rational efficiency is relatively high. This can be justified by the importance of transiting exergy. This exergy flow is embodied in the sea water used to cool sulfuric acid. So to increase the energy performance of the sulfuric unit we can think of a better use of energy continues in the sulfuric acid leaving the absorption towers.

The rational efficiency of the final tower is about 99%, however, its utilizable efficiency is about 63%. This is attributed to the importance of transit exergy embodied in the gas released to the atmosphere and the acid coming out at 86°C. Thus the best development of these two streams can improve the energy performance of the studied unit.

We note that the efficiency of the combustion furnace is about 70 %. However these losses are inevitable as they are due to the combustion reaction of sulfur which is an irreversible and highly exothermic reaction.

Table 5: Efficiencies of the main equipments.

Equipments	Rational efficiency	Utilizable efficiency
Blower	90,11	76,50
Drying tower	99,98	99,98
oven	70,78	69,33
Boiler	81,26	25,96
Superheater	97,54	66,53
Converter	99,00	99,00
GGH heat exchanger	99,50	89,33
GGC heat exchanger	97,04	67,24
Economizer 2	99,47	87,78
Economizer 1	93,16	78,62
I.Abs tower	98,84	77,42
F.Abs tower	99,82	63,08
Coolers	99,93	27,07

Internal exergy losses
Ex$_{int}$=78.63 MW

External exergy losses
Ex$_{ext}$=1.9MW

Exergy of products
EX$_p$ = 48.47 MW

Inlet exergy
Ex$_i$=178.42 MW

Exergy consumed
EX$_c$=129 MW

Outlet exergy
Ex$_e$= 50.37 MW
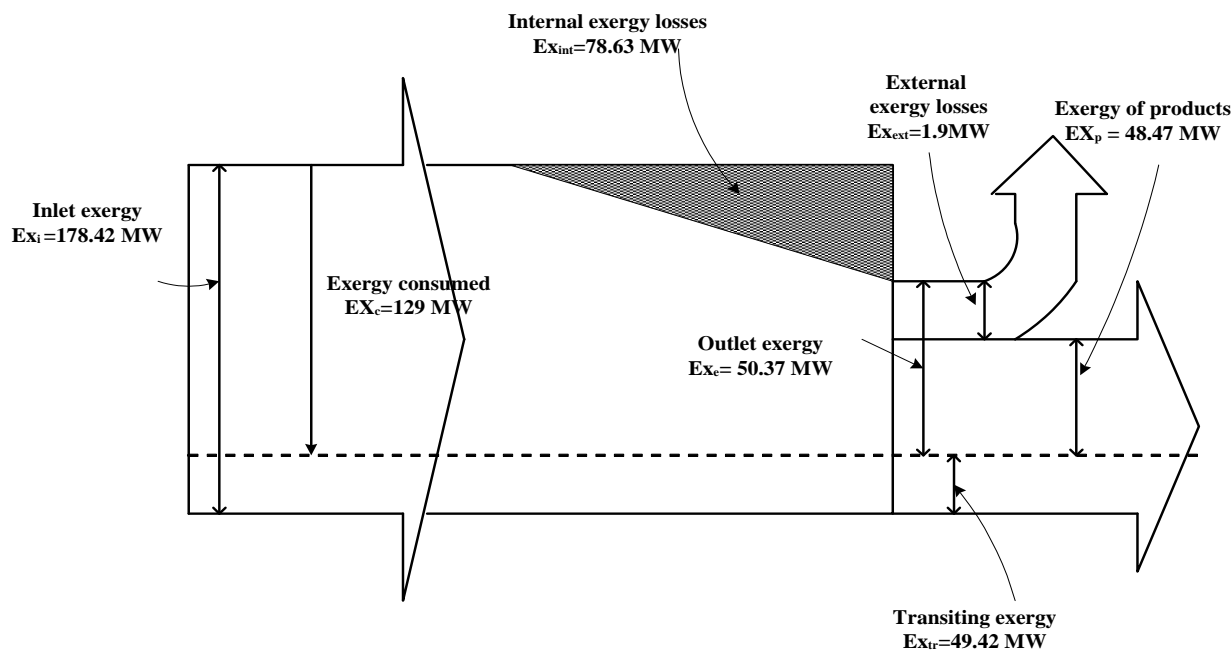
Transiting exergy
Ex$_{tr}$=49.42 MW

Figure.5: Exergy diagram of the sulfuric acid production unit.

4.3 Graphical presentation of the exergy balance

A graphical presentation of the exergy diagram of the sulfuric acid is given in Figure.5. It shows that the transiting exergy is about 50 MW, it correspond to about 27% of the total exergy input. This part of exergy is mainly embodied on the sea water used for the acid cooling. Thus to improve the overall efficiency of the studied unit we must think about using this part of exergy.

In the other hand the external losses represent 1.2% of the inlet exergy. This value is small relative to overall losses, but we can think of its reduce. Indeed this part of loss is embodied on the gas rejected to the atmosphere at 77°C. So the best handling of this flow reduce the exergy losses as well as the environmental thermal pollution.

Conclusion

In this study we have presented an exergy analysis of a sulfuric acid production unit. The overall efficiency was estimated to 56 % with a contribution by about 70% of the combustion step. The comparison of the rational and utilizable efficiencies shows the importance of the transiting exergy that represent about 27 % of the overall inlet exergy. This part of exergy is embodied in the sea water flow used for acid cooling. Thus the project of reducing the exergy losses should be oriented toward the management of the heat absorbed by sea water. In the same way a best recovery of the gas rejected to the atmosphere at 77°C will be an energetic and an environmental improvement.

List of abbreviations

Ex exergy, (kw)
F   molar flowrate, (kmol.s^{-1})

H   enthalpy, (kJ.kmol-1)
H$_0$ enthalpy at steady state, (kJ.kmol-1)
GGC gas- gas cold heat exchanger
GGH gas- gas hot heat exchanger
P   pressure, (Pa)
P$_0$ reference pressure, (Pa)
Q   heat duty, (W)
R   ideal gas constant, (J.K-1mol^{-1})
S   entropy, kJ.kmol^{-1}.K^{-1})
S$_0$ entropy at steady state, kJ.kmol^{-1}.K^{-1})
T   temperature, (K)
T$_0$ reference temperature, (K)
n   partial molar flowrate, (kmol.s^{-1})
x   molar fraction
W   work, (kW)
Indices
c   consumed
Ch chemical
D : destructed
e   outlet
ext external losses
i   inlet
int internal losses
j   compound
k   number of heat sources
l   number of work sources
m   number of compounds
p   product
ph physique
r   rational
tot total
tr   transiting
u   utilisable
Greek letters
γ   activity
Δ   Difference

ε Standard chemical exergy,(Jmol^{-1})

η efficiency

Φ fugacity coefficient

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