Exergy analysis in the Pulp and Paper Industry

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Abstract

The Pulp and Paper industry is the highest energy consumer in Portugal. This means that an exergy analysis is extremely relevant in order to better understand the real impacts of this industry and how it can reduce its environmental footprint. The pulp manufacturing process is divided into different stages, with an explanation of what happens in each one. This division has the goal of proposing a standardized way of making this type of analysis to this specific industry, so it becomes easier in future work to compare each stage of the whole process.

An exergy analysis is then performed to a pulp manufacturing process in 2002, specifically to the Kraft process used by The Navigator Company in Setúbal, Portugal. Based on Alexandre Martins' report from 2002, with the aid of an energy audit performed in 2014, it was possible to compute the exergy efficiencies for the digester, washing and black liquor evaporation process, the processing stage, and the recovery boiler and biomass co-generation process which were 41%, 36% and 66%, respectively. The overall efficiency is also calculated, presenting a very low exergy efficiency of 17.5% without taking into consideration the debarking and lime ovens stages, and 7% if all stages are considered.

Keywords: Exergy; Exergy analysis; Exergy efficiency; Pulp and Paper industry.

1. Introduction

Industries are the ones responsible for transforming the raw materials into new forms and assembling the final products to be delivered to the consumers whether it be the everyday people or other industries that need those products to produce their own. This sector is also liable for extracting the natural resources that they themselves need to achieve their final product. In Portugal, the biggest energy consumer industry is, by a relatively large margin, the pulp and paper production.

To meet the requirements of the world population, as well as help these industries have a better performance, some adjustments must be made regarding the usage of devices and processes that need energy to function properly. Efficiency is the optimum way to measure and improve these equipment or processes to assure that energy sources are not fully depleted before they can be replenished. There are two main types of efficiencies: energy efficiency and exergy efficiency, that result from either an energy or exergy analysis, respectively.

The first law of thermodynamics states that all energy is conserved, meaning it cannot be lost nor gained, only transformed or, in other words, the total energy of an isolated system remains the same. It is based on this law that energy analyses are made, with the embodied energy of a product being the result of the sum of all the energy required to produce it. Exergy analysis, besides the first law, are also based on the second law of thermodynamics which is related to the entropy of a system and the fact that it cannot decrease if the system is isolated and adiabatic. Due to the dependence on both laws, exergetic analysis are much more useful when comparing different systems.

By definition, exergy is the maximum theoretical useful work obtained until a system reaches its thermodynamic equilibrium with the environment. It can also be defined as the minimum theoretical work needed for a system to reach a certain state away from the equilibrium. There are certain characteristics about exergy that should be considered: when the environment is specified, a value can be calculated for the system in question, making exergy a property of that system; the value of exergy can never be negative since it results from a spontaneous process where the equilibrium can be reached with at least zero work; exergy is not conserved but it is destroyed by irreversibilities.

Exergy can be divided into three different kinds: mechanical, thermal or chemical. The definitions and differences between each can be found in the literature and other papers such as Ayres (2003), Buhler et al. (2016), or Sciubba and Wall (2007). Mechanical exergy is also known as kinetic or potential energy, whereas thermal exergy is related with heat. Chemical exergy is a little more complex, since it relates to the maximum theoretical work that could be developed in the chemical reaction that combines that substance with reactants present in the environment to produce substances that are in equilibrium with the environment (Ayres, 2003). When making an exergy analysis, it is also important to distinguish the exergy loss ($\dot{E}x_{loss}$) from the exergy destroyed ($\dot{E}x_{dest}$). $\dot{E}x_{loss}$ is represented by all the non-useful exergy that exits the system, typically by-products of the process that will not be used in any other step. On the other hand, $\dot{E}x_{dest}$ results from the difference between all the exergy entering the system and all the exergy exiting it and is related with the entropy increase in an irreversible process.

Finally, Gong (2005) made an exergy analysis to a pulp and paper mill in Sweden. The results for the energy and exergy efficiencies are displayed in Figure 1. In this specific mill, along with the sulfate pulp mill, there is also a chemi-thermomechanical pulp mill (CTMP) and a paper mill.

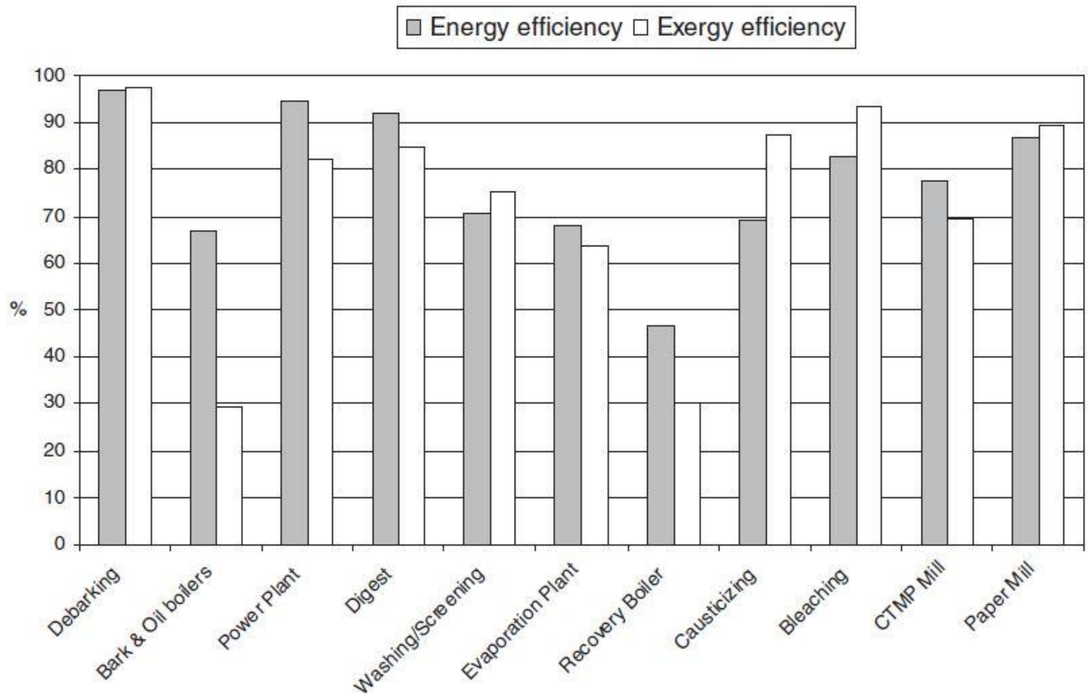

Figure 1 - Energy and exergy efficiencies for each process in a Swedish pulp and paper mill. Source: Gong (2005).

2. Case Study

This section will go over the reports from which most data was collected, Martins (2002) and an energy audit done in 2014. It will also include the method of analysis used to reach the final results, including how the control volumes should be divided, how they actually had to be divided and how to obtain the final exergy efficiencies for each process. Finally, a general a description for each process is given, as to better understand what is happening during the pulp manufacturing.

2.1 Alexandre Martins' report

The first part of this report revisits some of the work done before it, regarding other pulp and paper factories, followed by generic description of the Kraft process and its integration in the factory in order to understand how each part of the process works and the related mass and energy flows. Then, dividing it into smaller processes, the second part of the report describes in detail what is needed in each stage in terms of actual matter and energy requirements. The focus points are where heat is exchanged since it is possible to evaluate the current situation in the factory from this data. In this part, an analysis known as Pinch Analysis or *Análise do Ponto de Estrangulamento* (APE), in Portuguese, is also presented. This analysis provides crucial insight on the energy balances, optimizing the energy consumption of a given process based on thermodynamic principles. The third part of the report is where several options are presented to substitute different parts of the process by alternative ones that would result in a better usage of the energy produced in the factory itself. Final conclusions are then presented in the last section of the report, where the author gives his perspective on how his suggestions would be implemented and the conditions that have to be met for that to happen, discussing issues that may occur if in fact they follow through. Lastly, following this are several appendixes with some helpful information collected such as general energy consumption of the factory and more detailed tables of the heat exchanger components of each process.

2.2 Energy audit report

This document (Bento et al., 2016) follows a standard approach to this type of examinations, presenting tables, graphs and figures as well as moderately in-depth explanation of the entire pulp manufacturing process. The report starts by giving a description of the site history and how it operates nowadays, followed by a general characterization of the entire pulp process. The subsequent sections go into a comprehensive analysis of the energy consumption in each main part of the factory, finishing with the factory total consumption. In all these sub-sections we can find the same information: every step involved in this specific process, a table with its energy consumptions, such as electricity or steam, among others, where it can be improved. There is also a chapter about general services (electrical, compressed air and illumination) that account for all other aspects not mentioned previously. Finally, the authors leave some commentaries regarding energy saving options and opportunities that might help improve the facility consumption.

There are numerous differences between the data provided by Alexandre Martins' report and this one. The main difference is that the first one focusses on the heat exchange happening while the second merely points out the energy consumption for every segment. This means that there are practically no mass flows provided in the audit since they are not necessary for any of the calculations done in this type of report. The second major difference is how the data is presented. While Alexandre Martins disregards the energy with the enthalpy involved (in kW), the audit displays a table with the energy consumption in GJ, allowing an easy month by month analysis.

2.3 Method of analysis

Given the complexity of the pulp manufacturing process, it was necessary to divide it into several different control volumes. Setting the boundaries of each process is fundamental not only to understand what is happening at each stage, but also to evaluate any results that are calculated afterwards. As such, the entire process is divided into: Digester, Washing and Bleaching, Processing, Recovery and, finally, Biomass Co-generation.

The main reason why the information is and should be presented in this way is because each process has a defined purpose. The digester is where the raw material (wood/chips) is transformed into a pulp (brown in this case), with the black liquor produced as a by-product. This liquor will be very important later to produce high pressure steam, which will then be used to produce energy and other types of steam. From here, the pulp enters the washing and bleaching segment where the pulp will acquire the properties necessary to create high quality paper. Any nodes or not fully cooked segments of pulp will be returned to the digester, ensuring the best possible pulp characteristics while also wasting the least possible amount of wood. The processing is where all the pulp that was not sent to the paper mill will be dried, cut into sheets and stored. Parallel to all of this, we have the recovery boiler where the black liquor is concentrated and transformed into the green liquor while also producing high pressure steam. Finally, there is the biomass co-generation process where the high-pressure steam is used to produce electricity and medium and low-pressure steam flows. Not all parts of the factory where described, namely the preparation of wood and the causticizing and lime ovens. The first one simply debarks the eucalyptus and turns it into small chips, while the second one is where the green liquor is transformed into white liquor which will be used in the digester. The reason why the debarking of wood is not considered in this work is because there is no data regarding the main flows thus making an exergy analysis impossible to accomplish.

To obtain the most accurate results possible, the exergy was calculated using the data from both reports and other bibliographic materials such as Assari et al. (2014), Szargut (2007), Ptasinski et al. (2007). Moran et al. (2010) and Courchene et al. (2005). Since the audit has the most compact approach of all, the boundaries defined by it are: Digester, Washing and Black liquor evaporation, Bleaching, Processing and, lastly, Recovery Boiler and Biomass Co-generation. By using this approach, it was feasible to estimate many of the values such as liquors, condensates, electricity and even biomass, as well as mass flows, with some determined to close the mass balance of the process and others by collecting data from other sources, namely Assari et al.(2014). When it comes to the electricity, both reports give the electricity's specific energy consumption (SEC) for the entire factory, with the energy audit also providing it for each process. That being the case, it is possible to find how much electricity was consumed in 2002 for each process, by diving both values and obtaining the increase/decrease of electricity consumption, assuming all machinery and electrical appliances had this same increase/decrease in their efficiencies.

In order to obtain reasonable results, it was necessary to assume that the transportation of steam was adiabatic, meaning there is no heat loss and, since pressure remains constant along the pipes that carry this steam, temperature remained constant. This condition does not apply to every single flow, only to those with unknown temperatures. Another important aspect about steam flows is that the same process might, at different times, use different flows with different temperatures. As such, to compact the information as much as possible, the mass flows were compared and, in the case of being similar, a single flow with a weighted average mass flow and temperature was considered. Otherwise, if one of the mass flows was significantly higher than the others, the mass flow rate and temperature of that flow was considered. For the white liquor and condensates mass flow rates, the same temperature as the one presented in Assari et al. (2014) was used. It is also from this paper that most of the chemical exergy values were calculated. To do so, the physical exergy for each component was determined and then subtracted from the total exergy. This methodology was applied to all liquors (black, white, green and weak white), pulps (unbleached, bleached and white sheet) and chips. Regarding steams and condensates/water, the chemical exergy values were taken from Szargut (2007) and applied to these specific flows whereas the chemical exergy of biomass was taken from Ptasinski et al. (2007).

$$\dot{Q} - \dot{W} = \dot{Q}_{reaction} + \sum \dot{m}_{out}\Delta\bar{h}_{out} - \sum \dot{m}_{in}\Delta\bar{h}_{in} \quad (1)$$

In the energy balance (Equation 1), $\Delta\bar{h}$ represent the enthalpy variation (in regard to a reference temperature T_0), $\dot{m}_{in}$ and $\dot{m}_{out}$ are the mass flow rates coming into and out of the system, respectively, $\dot{Q}$ is the heat and $\dot{W}$ is the work. The heat that has to be provided (positive) or released (negative) for a reaction to occur, when the reactants and products are in the reference conditions, is denominated heat of reaction, $\dot{Q}_{reaction}$.

$$\dot{E}x_Q - \dot{E}x_W + \sum \dot{E}x_{in} - \sum \dot{E}x_{out} - \sum \dot{E}x_{dest} = 0 \quad (2)$$

Similarly, in the general exergy balance for steady-state conditions (Equation 2), $\dot{E}x_Q$ and $\dot{E}_W$ represent the exergy exchange in the boundaries of the system in the form of heat and work, respectively, where:

$$\dot{E}x_Q = \dot{Q}\left(1 - \frac{T_C}{T_H}\right) \qquad \dot{E}x_W = \dot{W} \quad (3), (4)$$

The exergy associated with the mass flows entering and exiting the system are given by $\dot{E}x_{in}$ and $\dot{E}x_{out}$. $\dot{E}x_{dest}$ is exergy that is destroyed during the process and cannot be avoided. T_C and T_H simply refer to the colder and hotter temperatures traded in the boundaries.

Regarding the physical exergy of a system (Equation 5), it is necessary to first calculate its enthalpy variation, Δh, and entropy variation, Δs. For the purposes of the work done here, Δh does not need to take pressure variation into consideration and Δs is only applied to liquids. Hence, Equations 6 and 7 can be used to produce the final results. The only exceptions are the Δh and the Δs of the steams involved during the process which can be found in Moran et al. (2010).

$$\dot{E}x_{ph} = \dot{m}\left(\Delta h - T_0\Delta s + \frac{v^2}{2} + gz\right) \quad (5)$$

$$\Delta h = h_1 - h_0 = c_p\,(T_1 - T_0) \qquad \Delta s = c_p\, ln\frac{T_1}{T_0} \quad (6), (7)$$

After the physical and chemical exergies are calculated, the final step is to add them in order to obtain the total exergy (Equation 8).

$$\dot{E}x_t = \dot{E}x_{ph} + \dot{E}x_{ch} \quad (8)$$

Finally, the exergy efficiency (ε) can be determined (Equation 9) using the previous equations and taking into consideration that the exergy output should only be of the useful components exiting each process.

$$\varepsilon = \frac{W_{max.output}}{W_{max.input}} = \frac{\dot{E}x_{t,out_{useful}}}{\dot{E}x_{t,in}} \tag{9}$$

2.4 Processes

The pulp and paper mill analyzed in this paper is in the industrial complex of the Miltrena Peninsula, in Setúbal and it is owned by The Navigator Company. Nowadays, the site is estimated to produce around 550 thousand tons of bleached eucalyptus kraft pulp per year, using a chemical process known as Kraft Process. The work here presented, however, reflects the process in 2002 since most of the data comes from that year. Figure 2 presents the five processes that occur during the manufacturing of paper pulp, along with the mass flows associated with each one of them.

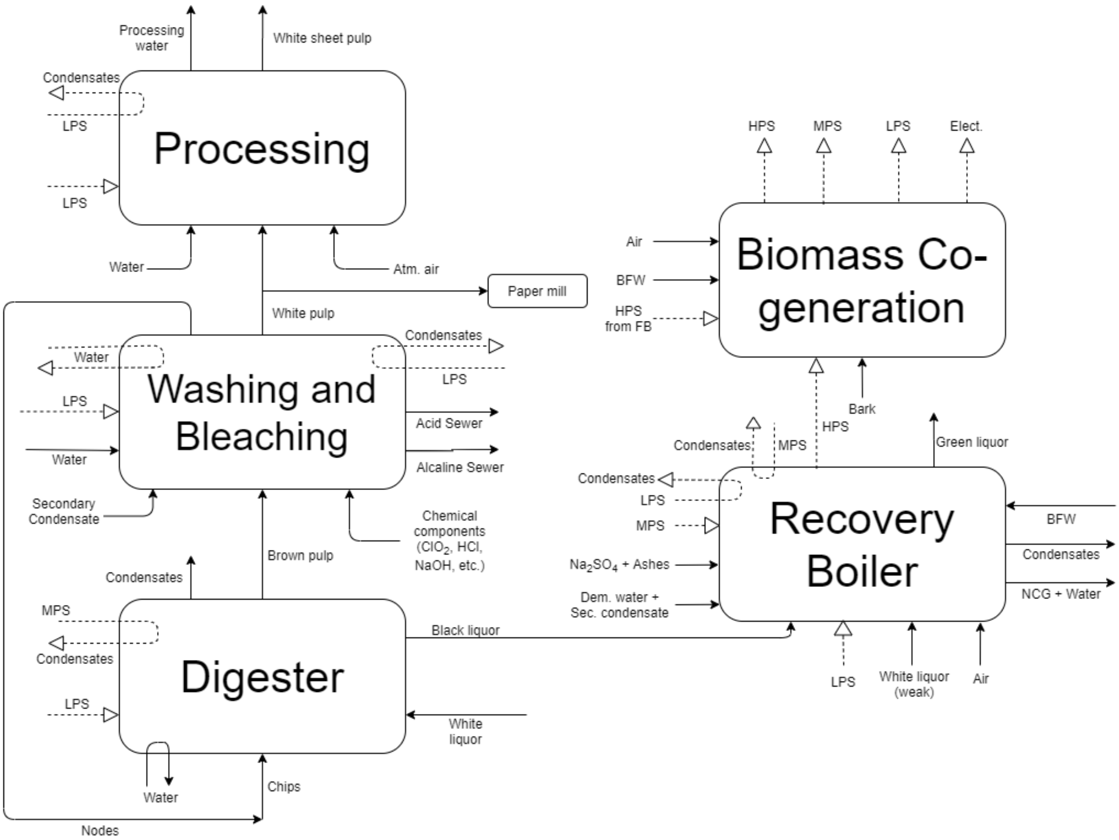

Figure 2 – Pulp process.

<u>Digester:</u> Low-pressure steam (LPS) pre-heats the wood chips that are later mixed with white liquor to dissolve the lignin and separate the fibers to form the brown pulp and black liquor is formed as a by-product. Medium-pressure steam (MPS) is used to re-heat this mixture along the process.

<u>Washing and Bleaching:</u> The brown pulp is washed with warm water in order to remove any remaining nodes that will be reintroduced in the Digester process. This washing process occurs in

counterflow, meaning while the brown pulp is going in one direction, the water is coming from the opposite one. Chemical components are then added to the brown pulp to give it the desired properties, including its characteristic whiteness.

Processing: Only 2/3 of the pulp leaving the previous process reaches this stage with the other 1/3 going to the paper factory itself. In the processing, the white pulp is washed and dried and then transformed into pulp sheets in order to be sold in the market or transported to other sites.

Recovery Boiler: The black liquor formed in the digester is concentrated through a series of evaporators. A combustion then occurs between the concentrated black liquor and the Na_2SO_4 and ashes that come from electrofilters, producing smelt and high-pressure steam (HPS) using the boiler feeding water (BFW). This smelt is then mixed with weak white liquor forming the green liquor.

Biomass Co-generation: Bark from the wood is burned to produce HPS. This, plus the HPS from the recovery boiler and, if necessary, a fuel-oil boiler (FB), are used in a turbine to produce MPS, LPS and electricity.

3. Results and discussion

In 2002, the production of brown pulp was about 1392 tons/day, whereas in 2014 this value was 1594 tons/day. This means there was, approximately, an 14.5% increase in brown pulp production between 2002 and 2014. The specific consumption of electricity in 2002 was 1.951 GJ/ton whereas in 2014 this value was 1.335 GJ/ton, meaning there was a decrease in the specific energy consumption of around 46%. Taking this value into consideration and knowing the specific consumption of electricity for each process in 2014, the electricity consumed in 2002 for each process was estimated.

For the energy balance of the digester, washing and black liquor evaporation process, presented in Table 1, the heat of reaction was taken from Courchene et al. (2005), where it was presented in $kcal/kg_{chips}$ and converted to kJ/s using the known mass flow rate of the chips.

Table 1 - Energy balance to Digester, Washing and Black liquor evaporation. (1) – from Courchene et al. (2005).

$\dot{m}\Delta h_{in}[MJ/s]$	$\dot{m}\Delta h_{out}[MJ/s]$	$\dot{W}[MJ/s]$	$\dot{Q}_{react}[MJ/s]$
154.2	141.5	-9.3	18.4(1)

Using Equation 1, we can see that the energy balance for the digester, washing and black liquor evaporation process is not closed. However, the gap represents approximately only 2.5% of $\dot{m}\Delta h_{in}$, which is a very small part of the overall energy involved during this process. This difference can be easily explained by the fact that not all the components are being taken into consideration, as explained earlier in this section

Regarding the exergy efficiencies, Table 2 presents them along with the lost and destroyed exergy for each process. The bleaching process is not represented since neither reports give any data regarding the mass flows of most of the main chemical components (NaOH, H_2O_2, SO_2, H_2SO_4 and O_2).

Table 2 – Exergy efficiencies and lost and destroyed exergy of all processes.

	ε[%]	$\dot{E}x_{loss}[MJ]$	$\dot{E}x_{dest}[MJ]$
Digester, Washing and Black liquor evaporation	41	14.3	429.4
Processing	36	22.2	26.5
Recovery Boiler and Biomass Co-generation	66	1.9	187.8
Aggregated	17.5	-	-
Aggregated w/ debarking and lime ovens and caustacizing	7	-	-

As shown, the least efficient process is the processing since it uses warm water (at high temperatures) as well as steam to wash the pulp and pre-heat the drying air, respectively. Other processes like the digester and recovery boiler also use steam, however it is typically done at lower temperature differences or use flash steam created during the process itself to help with the task of increasing the components temperatures. Regarding the $\dot{E}x_{loss}$ and the $\dot{E}x_{dest}$, the former is always smaller than the latter, with the processing having the lower difference. These values suggest that, in order to create a more efficient manufacturing process, ways to decrease $\dot{E}x_{dest}$ should be prioritized as well as creating alternatives in the processing stage.

4. Conclusions

Separating the stages into objective focused processes, where the boundaries are well defined, is the optimum way of achieving a better comprehension of the entire process while simplifying its multiple steps. Ultimately there would be a standardized way of dividing each industry so that it was possible to compare each step properly. The solution proposed here is the one that makes the most sense when discussing chemical pulping for paper production.

Regarding the Setúbal factory, the main conclusion that is drawn in this work is that the processing stage is the least efficient process studied, contrary to what is presented by Assari et al. (2014) and Gong (2005). In order to increase the overall efficiency of pulp mill, solutions should look into prolonging the time that each step takes, thus decreasing irreversibilities and, consequently, decreasing the exergy destruction. The exergy loss might be more difficult to decrease since most stages involve chemical reactions that produce by-products that cannot be avoided as they are part of the reaction itself.

Finally, energy audits should consider extending their data collection to include mass flows and temperatures of all the components involved in the process. In addition, the legislation should consider adding exergy consumptions to its reports and policies should change accordingly so that countries and businesses can make their decisions based on a more accurate read of their current circumstances.

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