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Definition of a Thermodynamic Parameter to Calculate Carbon Dioxide Emissions in a Catalytic Reforming Process

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Abstract

In the context of global warming, reduction of carbon dioxide emissions in oil and gas processes is an environmental and financial issue for process design and comparison. Environmental impact of a system can be determined by life cycle assessment (LCA). However this method presents limitations. Exergy is a thermodynamic function often chosen to complete LCA as it enables quantifying energetic efficiency of a process and takes into account the relation between the considered process and its environment. The aim of this work is to build a correlation between CO_2 emissions and a thermodynamic quantity which depends on exergy. For the process under consideration, this correlation has the following asset: it enables CO_2 emissions calculation without performing an LCA, when operating conditions are modified. The process studied here is naphtha catalytic reforming.

Keywords: life cycle assessment, exergy analysis, carbon dioxide, oil industry, catalytic reforming

1. Introduction

The development of industrial activities has induced environmental changes for several decades. To ensure a sustainable development, the environmental impact due to products and processes should be estimated. Intuitively a low environmental impact implies a high energy efficiency of the process under consideration. Life cycle assessment (LCA) is the most used method to calculate the environmental impact of a product or a process while exergy analysis gives information about the thermodynamic efficiency of a process. In this work, these tools are used together to outline a relation between CO_2 emissions and a thermodynamic quantity in order to determine a correlation for the catalytic reforming process. The first thermodynamic quantity tested is destroyed exergy in the process and the second quantity is a thermodynamic parameter including exergy of feed, product and utilities. The calculation of this thermodynamic quantity, for a process working under different conditions (temperature and feed modification for instance), enables then to know CO_2 emissions without performing a full LCA, which is a method with some limitations (listed in the next paragraph) and with very large boundaries making it unsuitable for process sizing and comparison.

2. Background

2.1. Life cycle assessment

Based on mass and energy balances, LCA is able to take into account inputs, outputs, and direct or indirect pollutants emissions related to the life cycle of a product, a process or a service (Chevalier, 1999). The scope of the analysis includes extraction and processing of raw materials, process construction and dismantling, product distribution, use, re-use, maintenance, recycling and final disposal. A set of ISO norms (ISO 14 040 - 14043, 2000) describes the four steps of LCA: definition of the goal and system limits, inventory analysis (data collection, mass and energy balances), impact assessment (impact classification and characterization and results aggregation), and improvement analysis (process conditions enhancement).

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So far, LCA has been mainly applied to products and use in process optimization has been quite rare (Burgess and Brennan, 2001-a). Azapagic (1999) has reviewed the emerging applications of LCA in process selection, design and optimization (multi-objectives optimization techniques) and Burgess and Brennan (2001-a) have reported applications of LCA to chemical processes. Lombardi (2003) has compared different technical solutions for CO_2 emissions reduction in power generation. Burgess and Brennan (2001-b) have outlined the importance of fugitive emissions in an LCA study on the desulphurization of petroleum gas oil. Arena et al. (2003) has compared three scenarios of solid waste management by a life cycle assessment study.

Despite the fact that LCA methodology has been standardized under the coordination of the Society of Environmental Toxicology and Chemistry (SETAC) (Consoli et al., 1993), it is often reported in the literature that LCA has not yet matured into a well defined tool (Udo de Haes, 1993). Indeed, there are some difficulties associated with the LCA methodology, for instance the definition of system boundaries. There are limitations regarding spatial and temporal scale. The choice to allocate co-products is not always straightforward and depends on the user (mass pro-rata, exergetic pro-rata, and substitution) (Bouvart and Prieur, 2006). Some impacts are difficult to assess. The use of fossil fuels and renewables can in general be well investigated; but hydropower, with its generally considerable impact on land use and nuclear power, with its potentially catastrophic impacts cannot be analyzed over their full scope. (Udo de Haes and Heijungs, 2007). Furthermore, LCA does not consider environmental sustainability of products or processes and ignores ecosystem services and products (Bakshi, 2002).

2.2 Exergy analysis

Exergy is a thermodynamic function that some have proposed to use to improve LCA as it enables studying the energy efficiency of a process and it takes into account the relation between the process under consideration and its environment. Exergy is more precisely based on the application of the first and second laws of thermodynamics and is a measure of energy quality. Szargut (2005) stated that the exergy of a material is the work or electrical energy necessary to produce that material in its specified state from materials common in the environment in a reversible way, heat being exchanged only with the environment. At full equilibrium between a system and an environment, the exergy of that system is equal to zero. Such a system state is called zero or dead state (Brodyansky et al., 1994).

Exergy only provides information about the current state of the system and its future ability to do work, but provides no information about the thermodynamic or energy history of the product or service in terms of ecological inputs (Bakshi, 2002).

Exergy of a system is divided into four components: kinetic exergy associated with relative motionİ potential exergy associated with gravitational or electromagnetic field differentials; physical exergy from pressure and temperature differentials; and chemical exergy arising from differences in chemical composition (Szargut, 2005; Kotas, 1995). In considering mass flows into and out of industrial processes, the first three components of exergy are negligible compared to chemical exergy (Ayres, 1998). To calculate the exergy of a mass flow rate, it is necessary to have information on the chemical composition of that stream compared to the environment in which it flows; the appropriate environmental reference state must also be characterized precisely. A general definition has been proposed by Szargut (2005): the reference state for most elements is taken to be either their most oxidized or chlorinated form. The composition of atmosphere, hydrosphere and earth's crust is taken as the reference state.

Physical exergy, B_{PH}, is the work that can be obtained by taking a given substance through reversible processes from its initial state (temperature T, pressure P) to the state determined by the environment conditions (T_0, P_0) and is given by the following expression:

$$\begin{aligned} B_{PH} &= (H - T_0 S) - (H_0 - T_0 S_0) \\ T_0 &= 298K \quad P_0 = 1bar \end{aligned} \tag{1}$$

Chemical exergy, B_{CH}, is the work that can be obtained by taking a substance at T_0 and P_0, to the state defined by the environmental reference composition. It represents exergy that nature has spent or should have spent to create the resource, respectively the waste. For a mixture of i components, its expression is given by the following expression:

$$\begin{aligned} B_{CH} &= \sum_i n_i \widetilde{b}_{ch,i} \\ &= \sum_i n_i (\mu_i - \mu_{i,0}) \end{aligned} \tag{2}$$

$$\widetilde{b}_{ch,i} = \widetilde{b}^0_{ch,i} + RT_0 \ln x_i \tag{3}$$

where $\widetilde{b}_{ch,i}$ is the partial molar chemical exergy of component i at temperature T_0 and pressure P_0 and corresponds to the difference between the partial molar Gibbs energy μ_i in the chemical state considered and in the chemical state of environment.

The exergy balance for open systems at steady state is given by:

$$\sum_{in}\left[\dot{W}+\dot{Q}\left(1-\frac{T_0}{T}\right)+\dot{B}_{fr}\right]=\sum_{out}\left[\dot{W}+\dot{Q}\left(1-\frac{T_0}{T}\right)+\dot{B}_{fr}\right]+\dot{B}_d \qquad (4)$$

The exergy content of the inputs is equal to the exergy content of the outputs (product or wastes) plus the exergy destroyed, $\dot{B}_d$, in the process. For the calculation of mass flow rate exergy, $\dot{B}_{fr}$, physical and chemical exergy are taken into account. Decreasing the destroyed exergy of a process means lower primary fuel consumption, thereby reducing the operating cost and increasing the process efficiency. This, in turn, will reduce emissions and wasted heat to the environment.

2.3. Exergy and environment

The most appropriate link between the second law and environmental impact has been suggested to be exergy, because it is a measure of the difference between the state of a system and the state of the environment (Szargut, 2005). The first systematic attempt to use exergy as a general quality measure of all resources, including renewables and mineral ores was conducted by Wall (1988). Indeed, resource degradation is a form of environmental damage because a resource is defined as a material, found in nature or created artificially, which is not in equilibrium with the environment, and that resource has exergy as a consequence of this disequilibrium (Rosen and Dincer, 2001). Waste exergy emissions can be viewed as a potential for environmental damage because they are not in stable equilibrium with the environment. Ayres (1998) stated that exergy provides a measure of the potential for emissions to cause environmental degradation because exergy is a measure of the distance of a product or a process from equilibrium.

Although the previous two points indicate simultaneously that in the environment, exergy in the form of resources is of value while exergy in the form of emissions is harmful, confusion can be avoided by considering or not the “constrained” exergy. Most of the resources found in the environment are constrained and are of value, in the sense that they stay in their state of disequilibrium in the long term, until they are used by man for a supposedly useful purpose, while unconstrained emissions of exergy are free to impact on the environment (Rosen and Dincer, 2001).

Finally, Cornelissen (1997) stated that exergy loss (both waste exergy emissions and internal exergy destructions) in a process can be used as a single criterion for the depletion of natural resources because it reflects the inefficiency of a process that implies a higher consumption of natural resources.

3. Methodology

The methodology, used in this work to evaluate carbon dioxide emissions, relies on combining LCA and exergy analysis to build a correlation for the naphtha catalytic reforming case. From the point of view of methodology, this process is interesting because carbon dioxide emissions are divided between direct emissions due to coke combustion in catalyst regeneration and indirect emissions due to electricity (compressors, pumps) and heat (combustion in furnaces) consumption.

This relationship is not universal and depends on the process and on the type of chosen utilities (electricity mix, heat production). The methodology is summarized in *Figure 1* and consists of the following steps:

- definition of system boundaries,
- process modeling,
- process simulation using *ProII 8.0* (Simsci, Lake Forest, United State of America) together with user added subroutines developed in *Fortran 77* to model reactors,
- life cycle assessment using *Simapro 6.0* (Pré Consultants, Amersfort, The Netherlands) to calculate CO_2 emissions,
- exergy analysis based on thermodynamic balances using a *Fortran* subroutine,
- comparison of a thermodynamic quantity and CO_2 emissions.

Finally a sensitivity analysis is conducted to evaluate the robustness of the relationship between CO_2 emissions and exergy.

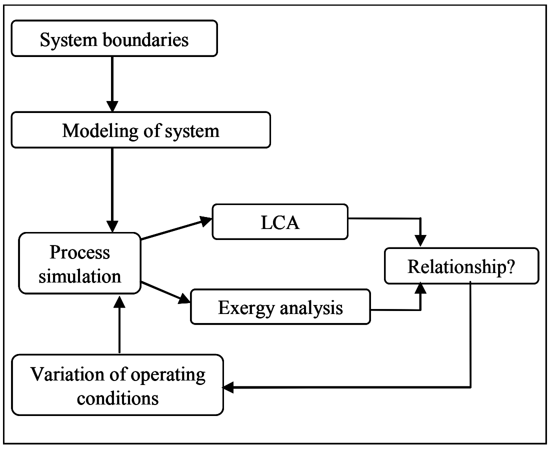

Figure 1. Methodology bloc scheme

4. Application

4.1. Naphtha catalytic reforming process

Catalytic reforming is a chemical process used to convert naphtha (of low octane number) produced during petroleum refining, into high-octane liquid products (Meyers, 1996). These are called reformates and are high-octane gasoline. Basically, the process "re-shapes" the hydrocarbon molecules of the naphtha feed and performs branching and aromatization. Alongside these reactions, cracking (breaking of large molecules into smaller ones) takes place.

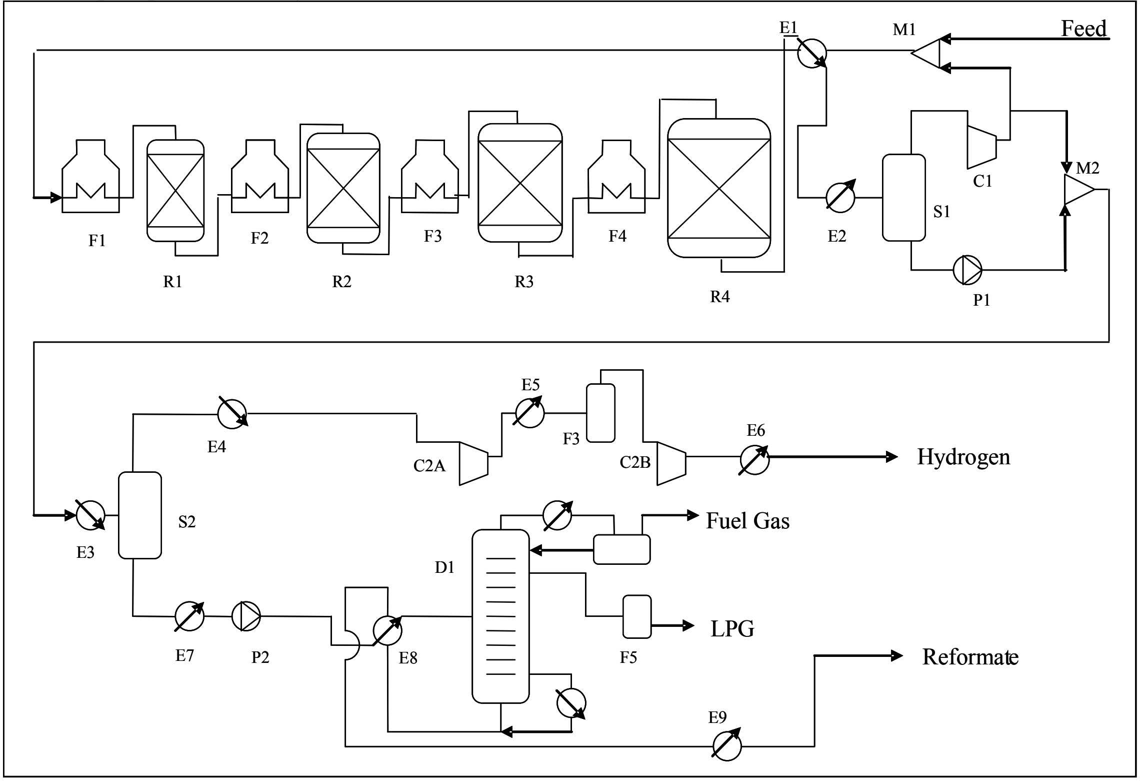

Figure 2. Process flow diagram of a continuous catalytic regeneration reforming process

The process produces very significant amounts of hydrogen gas and some light by-products. The process studied here is a typical continuous catalytic regeneration reforming process. This type of process (depicted in *Figure 2*) is characterized by a continuous *in situ* regeneration of the catalyst in a special regenerator. The reaction section is composed of four catalytic reactors and furnaces in series. Reactor inlet temperature is around 500°C and pressure is around 5 bar. Reactors are moving bed catalytic reactors with a Pt-Sn/Al_2O_3 catalyst. Flash distillations and an atmospheric distillation column enable separating products in the separation section. Catalyst regeneration is not represented on *Figure 2*.

4.2. Process modeling

The overall process is simulated with *ProII*. The gas/liquid equilibria are modeled according to the Grayson-Streed thermodynamic model included in *ProII*. That model is used for most refinery gas plant operations and hydrogen processes such as reforming and hydrocracking. The reference state for both liquid and vapor is the ideal gas and deviations from ideal behavior are determined by calculation of the fugacity coefficient for both phases.

Reactors are modeled with a *Fortran 77* subroutine because the simulator *ProII* does not achieve the modeling of catalytic reactors. The kinetic model used to represent chemical reactions taking place in reactors is based on the work by Van Trimpont, Marin and Froment (1981, 1988). The mechanism involves the following reactions: isomerization, paraffins ring closure, naphtenes dehydrogenation and hydrocracking. Plug flow is assumed in the reactors. Under these conditions, a mass balance for each component of the mixture and a global heat balance can be written. The differential equation system is solved by a Runge-Kutta method. Results have been validated by comparison with full scale industrial data.

5. Results

5.1. Simulations

The naphtha feeds used for simulations are presented in TABLE I. Naphtha 1 is mainly composed of naphtenes that can be easily converted into aromatics (which correspond to the best product to reach a high octane number) while Naphtha 2 is mainly composed of iso-paraffins that are easily cracked down.

TABLE I. FEEDS MOLAR COMPOSITION

	Naphtha 1 (%)	Naphtha 2 (%)
Paraffins	19	25
Iso Paraffins	24	32
Naphtenes	41	30
Aromatics	16	13

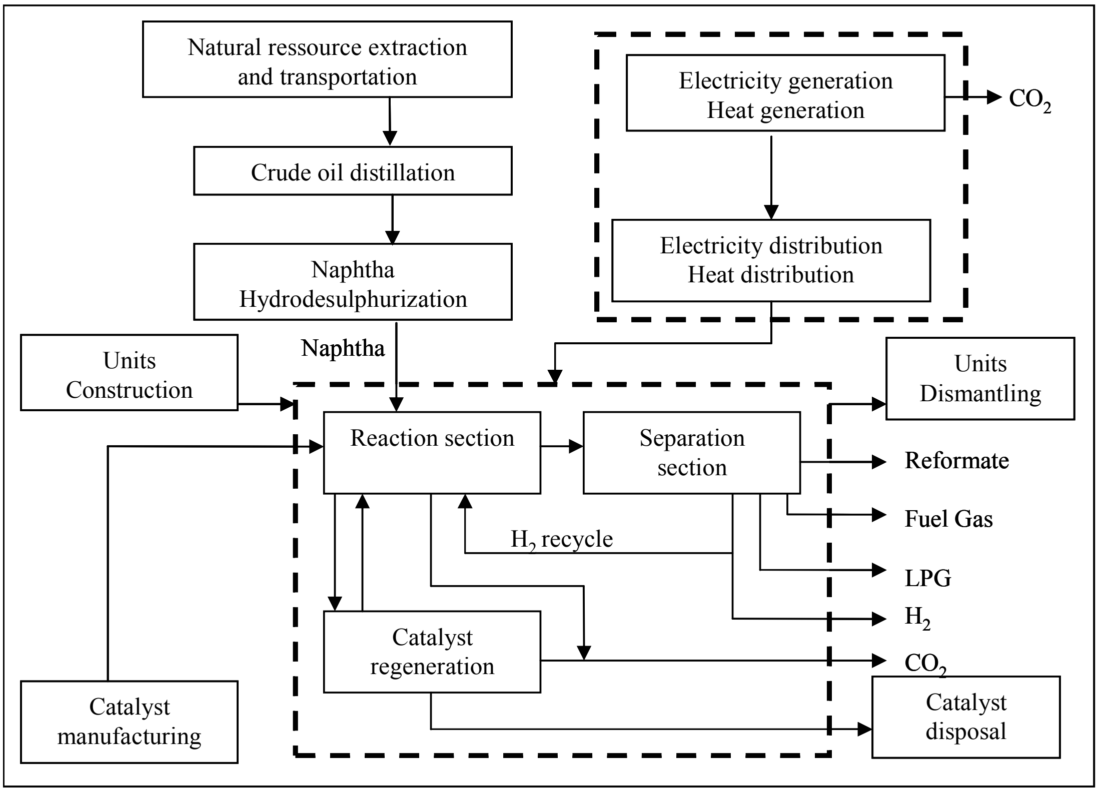

Figure 3. System boundaries for LCA

Simulations were run under different operating conditions. The first modified parameter is the reactor inlet temperature which was varied between 500°C and 550°C with 10°C increments (the same variation for each of the four reactors); this implies variation of energy provided by each intermediate fired heater. Because the temperature inlet increases, the reaction rates are modified. The second parameter modified is the type of feed. The methodology presented above is then applied to the reforming process for each temperature and each feed. This corresponds to twelve different scenarios.

5.2. Life cycle assessment

LCA enables calculating indirect CO_2 emissions (direct emissions due to catalyst regeneration is immediately calculated with *ProII*) involved by the process. Indeed, with the data base included in *Simapro*, indirect CO_2 emissions are related to parameters such as quantity of electric energy used in the process. The functional unit is the production of 1 kg of reformate. The system boundaries for this study and the links between processing units are shown in *Figure* 3. The areas delineated by dotted boundaries indicate steps that have been included in the LCA study. It includes a reaction and a separation section (also represented in *Figure 2*), a catalyst regeneration unit, and electricity and heat generation units. Natural resources extraction, transport operations and crude oil distillation have been excluded. This assumption is entirely valid when the same feeds are supplied to the system. In our case, the feed origins are not taken into account because we are only looking for CO_2 emissions immediately linked with catalytic reforming. This problem does not occur for electricity because the same electricity is provided in all cases. As stated in the literature (Lombardi, 2003), construction and dismantling phases of the units can be neglected and are therefore not taken into account. Concerning the inventory analysis step, the process model has been described previously and chosen parameters are: naphtha produced in European refineries, French (or European) electricity mix (with a large part of nuclear origin for the French one), and heat produced in natural gas furnaces (with low NO_X technology). The allocation mode chosen to take into account the three by-products (hydrogen, fuel gas and liquefied petroleum gas) is mass pro-rata. This allocation method is often chosen in literature (Burgess and Brennan, 2001-b). The CO_2 emissions results are allocated to reformate. The method used for impact assessment is Eco indicator 99. TABLE II summarizes the inventory data (provided by *ProII*) for the system considered for two scenarios with different feeds. Electricity and heat are given in energetic values.

TABLE II. INVENTORY ANALYSIS AND CO_2 EMISSIONS

	Naphtha 1, T=540°C	Naphtha 2, T=540°C
Resource use		
Feed [$t.h^{-1}$]	60.6	59.6
Electricity [MW]	4.1	3.8
Heat [MW]	23.7	22.0
Products		
H_2 [$t.h^{-1}$]	3.9	4.5
Fuel Gas [$t.h^{-1}$]	0.1	0.1
LPG [$t.h^{-1}$]	0.4	0.7
Reformate [$t.h^{-1}$]	56.2	54.2
Reformate CO_2 emissions [g.(reformate kg)$^{-1}$]	99.5	94.6
Indirect emissions	98.0	93.0
Heat	89.1	84.5
Electricity	8.9	8.5
Direct emissions	1.5	1.6

This table gives also the results in terms of CO_2 emissions and takes into account direct and indirect emissions. It is obvious that the treatment of Naphtha 2 implies less emissions than that of Naphtha 1 because of a lower utilities use. This smaller consumption can be explained by the fact that Naphtha 2 contains more iso-paraffins, which implies the enhancement of exothermic cracking reactions and therefore a lower consumption in furnaces.

The results depend for a large part on the choice of energy mix. In TABLE III, the comparison between the French electric mix and the European electric mix is made.

TABLE III. INFLUENCE OF THE ELECTRIC MIX ON CO_2 EMISSIONS

	Naphtha 1, T=540°C	Naphtha 2, T=540°C
Reformate CO_2 emissions - French electric mix [g.(reformate kg)$^{-1}$]	99.5	94.6
Indirect emissions	98.0	93.0
Heat	89.1	84.5
Electricity	8.9	8.5
Direct emissions	1.5	1.6
Reformate CO_2 emissions - European electric mix [g.(reformate kg)$^{-1}$]	125.9	118.9
Indirect emissions	124.4	117.3
Heat	89.1	84.5
Electricity	35.3	32.8
Direct emissions	1.5	1.6

As the French electricity is largely supplied by nuclear power plants, CO_2 emissions calculated with the French electricity mix are lower than CO_2 emissions calculated with the European mix.

5.3. Exergy analysis

Exergy analysis enables calculating exergy of streams (mass or heat fluxes) and the destroyed exergy in different units included in the overall process to find energetic inefficiency. The system considered is described in *Figure 2.* It should be noticed that the system considered is not exactly the same as for LCA. CO_2 emissions from heat and electricity generation have to be included in the environmental assessment because these utilities are directly used in the process. However, a usual power plant exergy analysis does not correspond to the scope of the study. Consequently, our exergy analysis is divided into two steps, the first is to calculate the destroyed exergy in the process (with a possible link with CO_2 emissions); the second is to find a thermodynamic parameter that enables taking into account heat and electricity generation taking place upstream without performing an exergy analysis for these utilities and considering exergy of feed and product. An exergy calculation tool has been built based on the previously described equations and integrated in *ProII*. Exergy analysis is summarized in TABLE IV for reactors working at an inlet temperature of 540°C for both feeds.

TABLE IV. EXERGY ANALYSIS

	Exergy Naphtha 1, T = 540°C	Exergy Naphtha 2, T = 540°C
Feed [MW]	745.9	736.5
H_2 [MW]	78.4	83.5
Fuel Gas [MW]	1.3	1.3
LPG [MW]	3.4	5.8
Reformate [MW]	667.7	646.9
Work [MW]	4.1	3.8
Heat [MW]	14.5	13.4
Destroyed exergy [MW]	13.8	16.1
Destroyed exergy [W.h.(reform. kg)$^{-1}$]	245.7	296.7

Work and heat calculated in TABLE II are net inlet values. An exergy balance applied on the process described in *Figure 2* gives a destroyed exergy of 13.8 MW for Naphtha 1 and 16.1 MW for Naphtha 2.

5.4. Comparison between CO_2 emissions and destroyed exergy

Results of simulations for different input temperatures are presented in *Figure 4*. This figure shows a positive non-linear link between CO_2 emissions allocated to reformate and destroyed exergy in the catalytic reforming process.

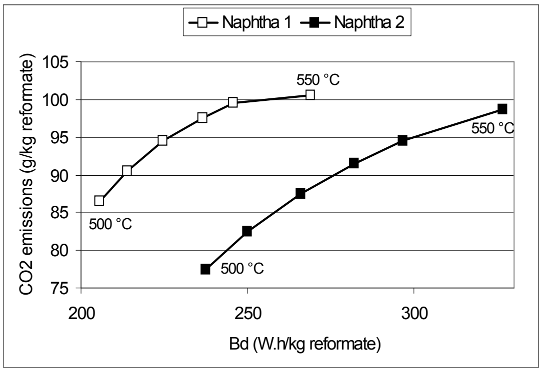

Figure 4. Evolution of CO_2 emissions as a function of destroyed exergy for Naphtha 1 and Naphtha 2

As temperature rises, CO_2 emissions increase less rapidly, while destroyed exergy increases steadily. This may be explained as follows. At high temperature (above 540°C), exothermic cracking reactions are promoted, reducing the need for heating in the furnaces, which implies a slow increase of CO_2 emissions. Moreover, cracking products increase destroyed exergy because of their low exergetic value. At low temperature (below 540°C), exergy consumed in furnaces increases sharply which implies an important increase of CO_2 emissions while exergy loss increases slowly.

At constant temperature, there is no positive relationship (for instance at 540°C, for Naphtha 1, $B_d = 205.8$ W.h.(kg$_{reformate}$)$^{-1}$ and $m_{CO2} = 86.5$ g.(kg$_{reformate}$)$^{-1}$ while for Naphtha 2, $B_d = 237.4$ W.h.(kg$_{reformate}$)$^{-1}$ and $m_{CO2} = 77.5$ g.(kg$_{reformate}$)$^{-1}$) because the compared products are not the same. Indeed, the octane number of reformate, which reflects the quality of reformate obtained, is 97.7 for Naphtha 1 and 96.1 for Naphtha 2 at a temperature of 540°C. It is important to notice that even if CO_2 emissions are low, the quality of the product obtained must also be taken into account. This link does not meet our requirements because destroyed exergy does not take into account quality of products and destroyed exergy in utilities production units is not considered.

5.5. Comparison between CO_2 emissions and a thermodynamic parameter

Since CO_2 emissions due to utility consumption are directly proportional to heat and electricity supplied to the process, to take into account the exergy of utilities is interesting. Moreover, the quality of feeds and products has also to be considered thanks to the exergy of these streams. The more feed is processed, the more utilities are needed. Therefore, utilities exergy is normalized by feed exergy. To take into account the product quality, the ratio is multiplied by the exergy of product. Consequently, the expression of the proposed parameter, I_p, including the exergy of utilities used in the process, $\dot{B}_{utilities}$, the exergy of product and feed stream, $\dot{B}_{product}$ and $\dot{B}_{feed}$, is given by the following expression:

$$I_p = \sum \dot{B}_{utilities} \frac{\sum \dot{B}_{product}}{\sum \dot{B}_{feed}} \qquad (5)$$

This expression enables avoiding the previous problem of system boundaries concerning utilities, as utilities exergy is considered instead of destroyed exergy to produce those utilities. Moreover, this ratio brings into play the quality of feed and product. A high I_p value means a high exergy of utilities but also a high quality of the products obtained. The plot of CO_2 emissions allocated to reformate as a function of I_p is given on *Figure 5*.

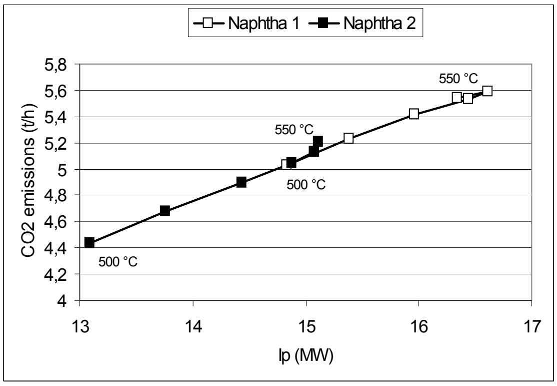

Figure 5. Evolution of CO_2 emissions in function of I_p for Naphtha 1 and Naphtha 2

This relationship shows an alignment of the Naphtha 1 and Naphtha 2 data points, confirmed by testing four other feeds (data not shown). This implies that the relation might be independent from the chosen feed. The quantitative relationship useful only for this process and for the chosen utilities is given by the expression:

$$\dot{m}_{CO2} = 0.3396 I_p \quad (6)$$

Carbon dioxide emissions are given in *t.h*$^{-1}$ and I_p is given in *MW*. This result would be different by changing the electric mix.

The parity diagram represented in *Figure 6* describes the plot of CO_2 emissions calculated by the previous correlation with respect to CO_2 emissions calculated from LCA. Results are evenly distributed along the bisecting line, meaning that CO_2 emissions are well represented by correlation (6). In the present case, the figure shows that the correlation is quite accurate.

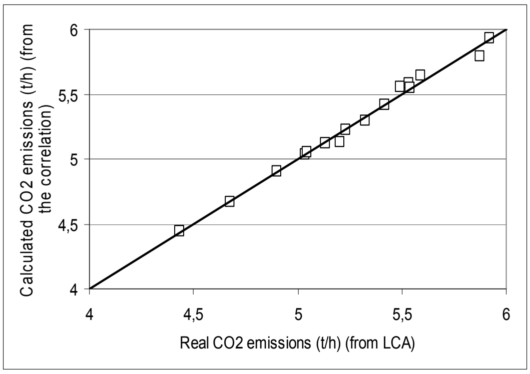

Figure 6. Parity diagram

Thus, when some operating conditions such as temperature and feed are changed in the process, the calculation of the thermodynamic parameter I_p estimates the CO_2 emissions, without performing a full LCA. The relationship enables checking which process conditions are the best from a CO_2 emissions point of view.

6. Concluding Remarks

LCA and exergy analysis are tools providing solutions to analyze processes and to determine environmental impact, energetic efficiency and quality of products. With these tools a relationship has been established between exergy destroyed and CO_2 emissions. In the discussed example, for a given feed, the first results show that the more exergy is destroyed, the higher the carbon dioxide emissions are. Given that this relationship was not independent from feed composition, another parameter has been established. This thermodynamic parameter is linked with CO_2 emissions with a mathematical correlation. LCA, which is a method well standardized but presenting some limitations and not very suitable for process design and comparison because of very large boundaries, can also be avoided.

The next objectives are to compare different versions of a process (for instance two different versions of the reforming process) in order to compare these processes from the point of view of CO_2 emissions and to validate theoretically the thermodynamic parameter.

Nomenclature

B	exergy
$\tilde{b}_{ch}$	partial molar chemical exergy
H	enthalpy
I_p	exergy indicator
m	mass
n	substance amount
P	pressure
Q	heat
R	ideal gas constant
S	entropy
T	temperature
W	work
μ	partial molar Gibbs energy

Subscripts

d	destroyed
i	component i
0	related to environment
in	inlet
out	outlet
fr	flow rate

Superscripts

0	standard
•	per time unit

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