Economic and environmental strategies for process design

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A B S T R A C T

This paper first addresses the definition of various objectives involved in eco-efficient processes, taking simultaneously into account ecological and economic considerations. The environmental aspect at the preliminary design phase of chemical processes is quantified by using a set of metrics or indicators following the guidelines of sustainability concepts proposed by IChemE (2001). The resulting multiobjective problem is solved by a genetic algorithm following an improved variant of the so-called NSGA II algorithm. A key point for evaluating environmental burdens is the use of the package ARIANE^{TM}, a decision support tool dedicated to the management of plants utilities (steam, electricity, hot water, etc.) and pollutants (CO_2, SO_2, NO, etc.), implemented here both to compute the primary energy requirements of the process and to quantify its pollutant emissions. The well-known benchmark process for hydrodealkylation (HDA) of toluene to produce benzene, revisited here in a multiobjective optimization way, is used to illustrate the approach for finding eco-friendly and cost-effective designs. Preliminary biobjective studies are carried out for eliminating redundant environmental objectives. The trade-off between economic and environmental objectives is illustrated through Pareto curves. In order to aid decision making among the various alternatives that can be generated after this step, a synthetic evaluation method, based on the so-called Technique for Order Preference by Similarity to Ideal Solution (TOPSIS) (Opricovic & Tzeng, 2004), has been first used. Another simple procedure named FUCA has also been implemented and shown its efficiency vs. TOPSIS. Two scenarios are studied; in the former, the goal is to find the best trade-off between economic and ecological aspects while the latter case aims at defining the best compromise between economic and more strict environmental impacts.

1. Introduction

In traditional chemical process design, attention has been focused primarily upon the economic viability. Yet, chemical plants can no longer be designed on the unique basis of techno-economic concerns and the other dimensions of sustainability – environmental and social – leading to the so-called "Triple Bottom line", must be part and parcel of the design phase. This study aims at the development of a design framework for eco-efficient processes, following the guidelines of the environmentally conscious design (ECD) methodology proposed by Allen and Shonnard (2002).

A major difficulty to tackle the problem is that there are many independent but often competing objectives that have to be considered simultaneously. Lots of ongoing researches aim at developing a set of metrics or indicators as proposed by IChemE (2001). In the dedicated literature, the amount of metrics may vary from 10 (AIChE, 1998) to 134 (CSD, 1996) to draw a quantitative profile of sustainability. The implementation of a process-oriented sustainability metrics has been carried out in parallel with the development of a design framework for eco-efficient processes, following the guidelines of the environmentally conscious design (ECD) methodology proposed by Allen and Shonnard (2002). For this purpose, several indexes of environmental impact including ozone depletion, Global Warming Potential, human and aquatic toxicity, photochemical oxidation as well as acid rain potentials have to be taken into account. Such problems lead to multiple and most often conflicting goals and must be solved by means of efficient multiobjective optimization tools. Many recent works used the combination of multiobjective optimization and life cycle assessment approach for the eco-design of chemical processes.

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Nomenclature

AP	Atmospheric Acidification Potential (eq t SO_2/y)
D_i	distillate flow rate in column of distillation i (kg/h)
EP	Eutrophication Potential (eq t PO_4^{3-}/y)
EB_i	ith environmental burden
F_i	alimentation flow rate (kg/h)
FUCA	French acronym for "Faire Un Choix Adéquat"
GWP	Global Warming Potential (eq t CO_2/y)
HDA	benzene production from toluene hydrodealkylation
Hi, Ho	enthalpies of input and output steams (kJ/kg)
HP	high pressure (Bar)
HTP	Human Toxicity Potential (t C_6H_6/y)
h_{Di}	enthalpy of distillate flow rate in device i (kJ/kg)
h_{Fi}	enthalpy of alimentation flow rate in device i (kJ/kg)
h_{wi}	enthalpy of waste flow rate in device i (kJ/kg)
ΔH_{stmi}	enthalpy of vapourization of water in unit i (kJ/kg)
ΔH_{stri}	enthalpy of vapourization of chemical stream i (kJ/kg)
LP	low pressure (Bar)
MCDM	multiple-criteria decision making
MILP	mixed integer linear programming
MINLP	mixed integer non linear programming
MP	medium pressure (Bar)
NLP	non linear programming
$\dot{m}_{FO}$	consumed flow rate of fuel oil (t/h)
$\dot{m}_{stmi}$	steam flow rate demand for device i (t/h)
$\dot{m}$	consumed flow rate of natural gas fuel (NM^3/h)
NSGA	non-sorted genetic algorithm
PCOP	PhotoChemical Oxidation or smog formation Potential (eq t C_2H_4/y)
PEI	potential environmental impact
Q_{bi}	heat supplied to reboiler i (kJ/h)
Q_{ci}	heat extracted from the unit i (kJ/h)
R	reflux ratio
ratio	fuel ratio (%)
SBX	simulated binary crossover
TOPSIS	Technique for Order Preference by Similarity to Ideal Solution
WAR	Waste Reduction Algorithm
W_i	waste flow rate (kg/h)
η	furnace or boiler yield (%)
ρ	correlation coefficient

This approach is increasingly being used in the literature (Azapagic & Clift, 1999; Berhane, Gonzalo, Laureano, & Dieter, 2009, 2010; Gonzalo, 2011).

This work aims at developing a design framework incorporating environmental issues during the preliminary stages of a process design. The environment component is identified as several design objective functions at the level of a process flow sheet synthesis. First, a brief literature survey dedicated to eco-friendly process design via modelling and optimization formulation is proposed in this paper. Then, the methodological framework adopted in this work for eco-efficient chemical process design is developed. In this study, optimization is performed by a genetic algorithm implemented in the Multigen library (Gomez et al., 2010) that turned out to be particularly well-suited for multiobjective process optimization. The well-known benchmark process for hydrodealkylation (HDA) of toluene to produce benzene (Douglas, 1988) is revisited in a multiobjective mode and illustrates the usefulness of the approach in finding environmentally friendly and cost-effective designs. A key point of the methodology is to capture in the modelling approach both process and utility production units, since the environmental impact of a chemical process is not only embedded in the products involved in the process but is also related to the energy consumption, the effect of flow recycle, percent conversion and so on. The trade-off between the seven considered objectives (production, annual cost and five environmental impacts) begins by a biobjective economic optimization (production, annual cost). Then, for a given production, the annual cost is deduced from the Pareto curve. Biobjective optimizations are implemented to establish multilinear relations between some objectives in order to reduce the multiobjective problem size to three antagonist criteria. Two cases are finally studied. In the former case study, under a fixed production level, the aim is to find the operating conditions for globally improving the environmental impacts. In the latter scenario, the objective is to satisfy environmental requirements as those that might be defined by an Environmental Protection in order to have a GWP (Global Warning Potential) value less than a given threshold; the other environmental impacts are required to be inferior to the values obtained in the first scenario.

2. Literature review related to eco-friendly process design

In recent years, eco-design, which consists in taking into account environmental assessment at the preliminary stage of process design, has been recognized as an efficient environmentally friendly alternative to traditional process design. Currently, there is no standardized methodology and almost no practical experience in integrating sustainable criteria into process design (Azapagic, Millington, & Collett, 2006). Generally, in this kind of problem, the objective is to simultaneously maximize profit while minimizing environmental impacts. Two approaches are generally considered in chemical system modelling, the first one is based on mathematical programming methods including either deterministic algorithms such as MINLP, NLP, MILP formulations or stochastic optimization techniques to solve this kind of problems. Some interesting references include: Surya and Alex (2002), Jia, Zhang, Wang, and Han (2006), Tveit and Fogelholm (2006), and Vasilios and Shang (2008). The second technique to model chemical processes is the use of simulators. Flowsheeting program packages, like CHEMCAD, Aspen Plus, HYSYS, PRO/II, and ProSimPlus, are commonly used in chemical engineering for process design. They can be used in an outer optimization loop to optimize different criteria. Some significant works can be found in Stanislav (2003), Lim, Dennis, Murthy, and Rangaiah (2005), Othman, Repke, Wozny, and Huang (2010), and Iskandar and Rajagopalan (2011). Carvalho, Gani, and Matos (2008) developed a generic and systematic methodology for identifying the feasible retrofit design alternatives of any chemical process. This systematic methodology has been implemented into an EXCEL-based software called Sustain-pro.

More generally, sustainable development takes into account the concept of life cycle assessment (LCA), which is a method for analysing and assessing the environment impact of a material, product or service throughout the entire life cycle. A whole life cycle includes all processes from the cradle to the grave, i.e. raw material, extraction, processing, transportation, manufacturing, distribution, use, reuse, maintenance, recycling and waste treatment. Björk and Rasmuson (2002) develop a new approach to design environmental impact as cumulative formation of ecopoints. They show that LCA is a valuable tool for environmental optimization of energy systems. The application of the whole life cycle assessment may be considered as very tedious due to the lack of information at some stage of its development. To circumvent this difficulties, a set of metrics or indicators following the guidelines of sustainability concepts have been developed, IChemE (2001), AIChE (1998), CSD (1996) and so on. These indicators use the concept of environmental

burdens defined as a quantitative measure of the potential contribution of substances released, to a particular environmental potential impact. It must be yet highlighted that they are often limited to a cradle-to-gate or gate-to-gate study. The environmental burdens are used to evaluate the environmental impacts in some strategies like the Waste Reduction Algorithm (WAR) (Chen & Li, 2008; Douglas & Heriberto, 1999; Heriberto, Jane, & Subir, 1999; Teresa, Raymond, Douglas, & Carlos, 2003), the IChemE sustainability metrics (Diniz da Costa & Pagan, 2006; Labuschagne, Brent, & van Erck, 2005) the Sustainable Process Index (SPI) (Ku-Pineda & Tan, 2006; Narodoslawsky & Krotscheck, 2004; Sandholzer & Narodoslawsky, 2007). For a given process, the potential environmental impacts are calculated from stream mass flow rates, stream composition and emissions from utility systems and a relative potential environmental score (index) for each chemical compound is deduced. The Sustainable Process Index (SPI) is an ecological evaluation system index specially developed for the requirements of process engineering. Narodoslawsky and Krotscheck (2004) used this environmental evaluation methodology to study the sustainability of energy production systems. The WAR algorithm is a methodology for determining the potential environmental impact (PEI) of a chemical process. The PEI balance is a quantitative indicator of the environmental friendliness or unfriendliness of a manufacturing process. The WAR algorithm was first introduced by Hilaly and Sikdar (1994). They introduced the concept of a pollution balance which was the precursor to the PEI balance. Douglas, Richard, and Heriberto (2000) used the WAR algorithm to evaluate the environment impact of an allyl chloride production facility. A systematic approach for sustainable assessment of chemical and energy production process which incorporates exergy analysis to quantify the efficiency of a process and an enhanced inherent safety index to quantify the societal impact of a process, has been proposed by Li, Zanwar, Jayswal, Lou, and Huang (2011). According to Chen and Li (2008), this method generally divided the impact categories into two general areas with four categories. The first general area is the global atmospheric level involving Global Warming Potential (GWP), Ozone Depletion Potential (ODP), Acidification and acid-rain Potential (AP), PhotoChemical Oxidation or smog formation Potential (PCOP). The second general area is related to the local toxicological impact level and is associated with Human Toxicity Potential by Ingestion (HTPI), Human Toxicity Potential by either inhalation or dermal Exposure (HTPE), Aquatic Toxicity Potential (ATP) and Terrestrial Toxicity Potential (TTP). Like the WAR algorithm, the methodology developed by IChemE (2001), also uses the concept of potency factors for different types of pollutants. The total environmental burden related to an environmental impact category is calculated by summing the product of different potency factors of pollutant with the mass flow rate of pollutants which contribute to this environmental burden. Vasilios and Shang (2008) used the IChemE (2001) method to calculate the environmental impacts of utility production systems. This technique is also adopted in this study for the environmental evaluation, since these criteria have been identified as representative of the process industries. At the final stage of the optimization step, it may be necessary to help decision makers in determining trade-off solutions, several decision analysis methods can be implemented. Pirdashti, Ghadi, Mohammadi, and Shojatalab (2009) and Zhou and Poh (2006) classify decision analysis methods into three main groups: single objective decision making (SODM) methods, Multi-Criteria Decision Making (MCDM) methods, and decision support systems (DSS). Their analysis highlights that MCDM methods that are structural approach to analyse problems with several criteria and alternatives are the most widely used strategies. They help decision makers to make consistent decisions by taking all the important objective and subjective factors into account. A comparison can be found in Gough and Ward (1996). Among the available techniques, the so-called Technique for Order Preference by Similarity to Ideal Solution (TOPSIS) which belongs to MCDM group is used for identifying the set of optimal parameters in numerous investigations. Li, Zhang, Zhang, and Suzuki (2009), Jia et al. (2006), Ren, Zhang, Wang, and Sun (2007) used TOPSIS as multicriteria decision method to trade-off solutions.

3. Eco-efficient chemical process design

3.1. General framework

The design of eco-efficient chemical processes proposed in this study (see Fig. 1) integrates a mathematical model for the considered process, coupled with an impact assessment model, which is embedded in an outer multiobjective optimization loop. For process modelling, commercial design and flowsheeting packages could be classically used. The proposed framework is an alternative design methodology for waste minimization to the so-called WAR algorithm (Cabezas, Bare, & Mallick, 1999; Young, Scharp, & Cabezas, 2000), which has been extensively used in the literature. Let us recall that this method is based on a potential environmental impact (PEI) balance for chemical processes. The PEI is a relative measure of the potential for a chemical to have an adverse effect on human health and the environment (e.g. aquatic ecotoxicolgy, global warming, etc.). The result of the PEI balance is an impact (pollution) index that provides a quantitative measure for the impact of the waste generated in the process.

Recently, several systematic methodologies have become available for the detailed characterization of the environmental impacts of chemicals, products, and processes, which include Life Cycle Assessment. The idea is to use their potential to develop a Life Cycle Analysis method dedicated to process development. Since our approach focuses on decreasing the environmental impacts of the manufacturing stage and utility systems, only a "cradle-to-gate" analysis is performed.

A key point of the methodology implemented here is the use of ARIANE™ (http://www.prosim.net/en/energy/ARIANE.html, 2005) a decision support tool dedicated to the management of plants that produce energy under the form of utilities (steam, electricity, hot water, etc.) included in the Plessala™ module developed by ProSim S.A. (2005). ARIANE™ is used here both to compute the primary energy requirements of the process and to quantify the pollutant emissions due to energy production.

3.2. Multiobjective optimization

Like many real world examples, the problem under consideration involves several competing measures of performance, or objectives (Collette & Siarry, 2002). Using the formulation of multiobjective constrained problems of Fonseca and Fleming (1998), a general multiobjective problem is made up a set of n criteria f_k, $k = 1, \ldots, n$, to be minimized or maximized. Each f_k may be nonlinear, but also discontinuous with respect to some components of the general decision variable x in an m-dimensional universe U.

$$f(x) = (f_1(x), \ldots, f_n(x)) \tag{1}$$

This kind of problem has not a unique solution in general, but presents a set of non-dominated solutions named Pareto-optimal set or Pareto-optimal front. The Pareto-domination concept lies on two basic rules: in the universe U a given vector $u = (u_1, \ldots, u_n)$ dominates another vector $v = (v_1, \ldots, v_n)$, if and only if,

$$\forall i \in \{1, \ldots, n\} : \quad u_i \leq v_i \wedge \exists\, i \in \{1, \ldots, n\} : \quad u_i < v_i \tag{2}$$

For a concrete mathematical problem, Eq. (2) gives the following definition of the Pareto front: for a set of n criteria, a solution $f(x)$, related to a decision variable vector $x = (x_1, \ldots, x_m)$, dominates

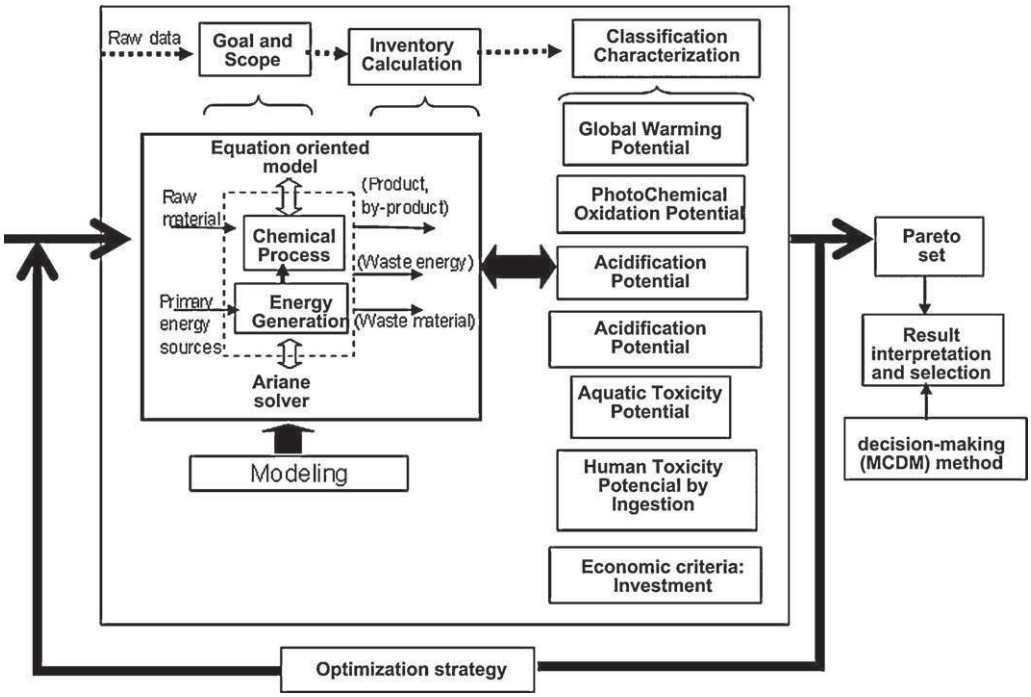

Fig. 1. General optimization and modelling framework.

another solution $f(y)$, related to $y=(y_1,\ldots,y_m)$ when the following condition is checked (for a minimization problem),

$$\forall i\in\{1,\ldots,n\}:\quad f_i(x)\le f_i(y)\wedge\exists\, i\in\{1,\ldots,n\}:\quad f_i(x)<f_i(y) \tag{3}$$

On a given set of solutions, it is possible to distinguish non-dominated sets. The last definition concerns the Pareto optimality: a solution $x_u\in U$ is called Pareto-optimal if and only if there is no $x_v\in U$ for which $v=f(x_v)=(v_1,\ldots,v_n)$ dominates $u=f(x_u)=(u_1,\ldots,u_n)$. These Pareto-optimal non-dominated individuals represent the solutions of the multiobjective problem.

Faced to the diversity of mathematical problems, it is recognized that there is not a unique and general algorithm able to solve all the problems perfectly. Actually, method efficiency for a particular example is hardly predictable, and the only certainty we have is expressed by the No Free Lunch theory (Wolpert & Macready, 1997): there is no method that outdoes all the other ones for any considered problem. This feature generates a common lack of explanation concerning the use of a method for the solution of a particular example, and usually, no relevant justification for its choice is given a priori. A possible solution was to develop several algorithms, distinguishing them by their structure and by their type of variables (continuous, integer, binary) and collect them into a database: Multigen (Gomez et al., 2010) lies on this principle, and currently, six different algorithms are available. The aim was to treat multiobjective constrained-optimization problems involving various types of variables (boolean, integer, real) and some of these problems can be structural optimization ones. Multigen has been implemented in VBA and interfaced with Microsoft Excel®. The algorithms must handle constraints as well as Pareto domination principles. In that way, procedures based on independent objectives to carry out the selection (like VEGA, Schaffer, 1985) are not adapted to the considered problems. Procedures based of the niche notion (NPGA – Horn, Nafpliotis, & Goldberg, 1994; MOGA – Fonseca & Fleming, 1993) cannot guarantee a correct convergence of the Pareto front, due to the low diversity of the generated populations. Methods like SPEA (Zitzler & Thiele, 1999) and NSGA II (Deb, Pratap, Agarwal, & Meyarivan, 2002) favour not dominated isolated individuals. In SPEA, the probability of selection is a function of the individual isolation, which is quite difficult to implement. In NSGA, individuals from the most crowded zones are eliminated according to a crowding sorting. Taking into account all the previous items, NSGA II was chosen as the basis of development of the Multigen library. In this paper the package NSGA IIb, which differs from the initial NSGA II by the crossover operator, has been retained. Note that, NSGA IIb implements the same algorithm than NSGA II, with corrections on crossover operator to avoid the creation of clones inherent of SBX original version. When the generated random number used to perform the crossover is greater than a given crossover probability, the crossover may produce two children identical to the parents: SBX crossover coded in NSGA IIb includes a forced mutation of children when this event occurs. The objective is to avoid unnecessary calculations for clones of existing solutions: all solutions generated by the reproduction procedure are statistically different. The NSGA IIb algorithm was implemented to treat variables of different nature, either continuous, or integer. Since our analysis is restricted to treat continuous variables, the way to handle integer variables is not detailed in this paper. The interested reader will fin more information in (Gomez et al., 2010).

Constrained multiobjective optimization is the most common, kind of problem in engineering applications. In general, three kinds of constraints are considered: simple inequality ($\le$), strict inequality ($<$), and equality ($=$):

$$\left.\begin{array}{l} g(x)\le c1\\ r(x)<c2\\ h(x)=c3 \end{array}\right\}\Leftrightarrow\left\{\begin{array}{l} constr_1(x)=c1-g(x)\ge 0\\ constr_2(x)=c2-r(x)>0\\ constr_3(x)=c3-h(x)=0 \end{array}\right. \tag{6}$$

where (g,r,h) are real-valued functions of a decision variable $x=(x_1,\ldots,x_m)$ on an m-dimension decisional search space U, and $(c1, c2,$

$c3$) are constant values. In the more general case, these constraints are written as vectors of the type:

$$\begin{aligned}
\vec{constr}_1(x) &= ((c1 - g(x))_1, \ldots, (c1 - g(x))_{n1}) \\
&= (constr_1(x)_1, \ldots, constr_1(x)_{n1}) \geq 0 \\
\vec{constr}_2(x) &= ((c2 - r(x))_1, \ldots, (c2 - r(x))_{n2}) \\
&= (constr_2(x)_1, \ldots, constr_2(x)_{n2}) > 0 \\
\vec{constr}_3(x) &= (-|(c3 - h(x))|_1, \ldots, -|(c3 - h(x))|_{n3}) \\
&= (constr_3(x)_1, \ldots, constr_3(x)_{n3}) = 0
\end{aligned} \tag{7}$$

where $n1$, $n2$, and $n3$ are respectively, the number or inequality, strict inequality and equality constraints. This formulation implies that each $constr_i$ value will be negative if and only if this constraint is violated. The conversion of Eq. (6), that is a classical representation of constraints set, to Eq. (7) representation constitutes the first step of a unified formulation of constrained-optimization problems. In practice, due to round-off error on real numbers, the equality constraint $constr_3$ was modified as follows:

$$\begin{aligned}
&\vec{constr}_3(x)' = (-|(c3 - h(x))|_1 + \varepsilon_1, \ldots, -|(c3 - h(x))|_{n3} + \varepsilon_{n3}) \\
&= (\vec{constr}_3(x)) + \vec{\varepsilon} = 0 \\
&\vec{\varepsilon} = (\varepsilon_1, \ldots, \varepsilon_{n3}), \quad \forall i \in \{1, \ldots, n3\}, \quad \varepsilon_i \in R
\end{aligned} \tag{8}$$

$\vec{\varepsilon}$ is called a "precision vector" of the equality vector, and takes low values (less than 10^{-6} for example). This approximation is not necessary when equality constraint involves only integer or binary variables.

From Eqs. (7) and (8), the constraint satisfaction implies the maximization of violated constraints in vectors $constr_1$, $constr_2$, and $constr_3$. According to Fonseca and Fleming (1998), the satisfaction of a number of violated inequality constraints is, from Eq. (7), a multiobjective maximization problem. From a theoretical point of view, a constrained multiobjective optimization problem can be formulated as a two-step optimization problem. The first step implies the comparison of constraint satisfaction degrees between two solutions, using the Pareto's domination definition of Eq. (3), but a more simple solution consists in comparing the sum of values of violated constraints only, as in NSGA II algorithm of Deb et al. (2002), which implies there are no priority rules between constraints.

3.3. Multiple-criteria decision making (MCDM)

3.3.1. Introduction

MCDM approaches are major parts of decision theory and analysis. MCDM are analytic methods to evaluate the advantages and disadvantages of alternatives based on multiple-criteria. The objective is to help decision-makers to learn about the problems they face, and to identify a preferred course of action for a given problem. Huang, Poh, and Ang (1995) mentioned that decision analysis (DA) was first applied to study problems in oil and gas exploration in the 1960s and its application was subsequently extended from industry to the public sector. Till now, MCDM methods have been widely used in many research fields. Different approaches have been proposed by many researchers, including single objective decision-making (SODM) methods, MCDM methods, and decision support systems (DSS). Literature shows that among MCDM methods, DA strategies are the most commonly used (Zhou & Poh, 2006). The selection of a single Pareto point from the Pareto frontier may be difficult as the number of objectives increases. Some intuitive methods such as the so-called "knee-method" could be efficient for a binary case. This is why other methods are necessary to tackle the multicriteria nature of the results generated by the GA.

3.3.2. Finding knees

Branke, Deb, Dierolf, and Osswald (2004), and Taboada and Coit (2006) suggest picking the knees in the Pareto front, that is to say, solutions where a small improvement in one objective function would lead to a large deterioration in at least one other objective.

3.3.3. TOPSIS

In practice, the Technique for Ordering Preference by Similarity to Ideal Solutions (TOPSIS method) and other multiple attribute decision making (OMADM) methods are the most popular (Ren et al., 2007; Yoon & Hwang, 1995). In this work, the TOPSIS methodology was used as decision making tool. After the generation of the Pareto-optimal set, the TOPSIS method is used to aid decision–maker in trade-offing the whole alternatives. The basic principle of TOPSIS is that the chosen alternative should have the shortest distance from the ideal solution and the farthest distance from the negative-ideal solution (Opricovic & Tzeng, 2004; Ren et al., 2007; Saaty, 1980).

Steps used in TOPSIS as described by Ren et al. (2007) are briefly described in what follows:

Step 1: All the original criteria receive tendency treatment. That consist in transforming the cost criteria into benefit criteria, which is shown in detail as follows:

(i) The reciprocal ratio method ($X'_{ij} = 1/X_{ij}$) refers to the absolute criteria;
(ii) The difference method ($X'_{ij} = 1 - X_{ij}$), refers to the relative criteria.

After tendency treatment, construct a matrix

$$X'[X'_{ij}]_{n\times m}, \quad i = 1, 2, \ldots, n; \quad j = 1, 2, \ldots, m. \tag{9}$$

Step 2: Calculate the normalized decision matrix A as follows:

$$A = [a_{ij}]_{n\times m}$$

$$a_{ij} = \frac{X_j'^i}{\max(X_j'^i)} \quad (j = 1, 2 \ldots, m.) \quad \text{for a criterion to be maximized} \tag{10a}$$

$$a_{ij} = 1 - \frac{X_j'^i}{\max(X_j'^i)} \quad (j = 1, 2 \ldots, m.) \text{ for a criterion to be minimized} \tag{10b}$$

Step 3: Determine the positive ideal and negative ideal solution from the matrix A

$$A^+ = (a_{i1}^+, a_{i2}^+, \ldots, a_{im}^+), \quad a_{ij}^+ = \max_{1\leq i\leq n}(a_{ij}), \quad j = 1, 2, \ldots, m \tag{11}$$

$$A^- = (a_{i1}^-, a_{i2}^-, \ldots, a_{im}^-), \quad a_{ij}^- = \min_{1\leq i\leq n}(a_{ij}), \quad j = 1, 2, \ldots, m \tag{12}$$

Step 4: Calculate the separation measures, using the n-dimensional Euclidean distance. The separation of each alternative from the positive ideal solution is given as

$$D_i^+ = \sqrt{\sum_{j=1}^{m} W_j(a_{ij}^+ - a_{ij})^2} \tag{13}$$

Similarly, the separation from the negative ideal solution is given as

$$D_i^- = \sqrt{\sum_{j=1}^{m} W_j (a_{ij}^- - a_{ij})^2} \quad (14)$$

Step 5: For each alternative, calculate the ratio R_i as

$$R_i = \frac{D_i^-}{D_i^- + D_i^+}, \quad i = 1, 2, \ldots, n \quad (15)$$

Step 6: Rank alternatives in increasing order according to the ratio value of R_i in step 5

3.3.4. FUCA

FUCA is the French acronym for Faire Un Choix Adéquat (Make An Adequate Choice). This simple method is based on individual rankings of objectives; for a given criterion, rank one is assigned to its best value and rank n (n being the number of points of the Pareto front) to the worst one. Then, for each point of the front, a weighted summation (the weights representing the preferences) of ranks is performed, and the choice is carried out according to the lowest values of the sum. In a recent paper (Moralez-Mendoza, Perez-Escobedo, Aguilar-Lasserre, Azzaro-Pantel, & Pibouleau, 2011) the FUCA method was compared with classical MCDM procedures on a tricriteria problem related to the portfolio management in a pharmaceutical industry. For each solution found by ELECTRE (Teixeiro de Almeida, de Miranda, & Cabral Seixas Costa, 2004), PROMETHEE (Zhaoxa & Min, 2010) and TOPSIS, the FUCA ranking is also reported. A very good agreement between the three classical MCDM methods and FUCA can be observed, showing the efficiency of the FUCA procedure, which always finds the best solution selected by one of the others.

3.4. Utility production modelling

ARIANE^{TM} software tool has been developed by ProSim Company (French chemical engineering Software Company) for designing assistance and optimal operation of power plants. ARIANE^{TM} makes it possible to optimize and model any energy combined production plant (steam, hot water, electricity, compressed air, cooling), whatever its size and complexity. ARIANE^{TM} is a decision support tool dedicated to the management of plants that produce energy under the form of utilities (steam, electricity, hot water, etc.). ARIANE^{TM}, written in VBA, presents a standard library of unit operations involved in power plants: boilers (mono-fuel, bi-fuel, electrical), turboalternators (backpressure turbines, sidestream turbines, condensating turbines), switchable or permutable turbines (in parallel with electric motors), fuel turbines and thermal engines (with or without recovery heat exchanger, with or without post combustion boiler), valves, heat exchangers and deaerators. All these unit operations can be described, from the simplest (minimalist modelling, with default values) to the most complex (very fine modelling of the characteristics and technical constraints); the complexity of physical models requiring in many cases to use nonlinear equations. The inherent model complexity and its associated equations are totally transparent for the user. In the framework of eco-design modelling, the problem lies in integrating environmental aspects inside ARIANE software, so that emissions can be taken into account in the global energy balance. Estimation methods, to evaluate the emissions of all involved power plants equipment items have then to be compatible with existing routines that not require numerous and complex input data. In ARIANE, classical pollutants as nitrogen oxides, carbon and sulphur oxides, and also solid particles like dusts, are well known.

The emissions modelling process starts from the calculation of pollutant amounts in the smokes of each equipment item, global emission flow rate is finally the sum of all flow rates, for all site smoke emitters. Another problem lies then in the accurate modelling of the combustion phenomena in all equipment items of a power plant. We give below some of the equations of units operations implemented in ARIANE^{TM} and used at the eco-efficient modelling stage.

4. Application to HDA (hydrodealkylation of toluene) process

4.1. Overview of the HDA process

The traditional method of manufacturing benzene from the distillation of light oils produced during the manufacturing of coke has been overtaken by a number of processes: The sources are now: catalytic reforming, hydrodealkylation of toluene (HDA) and toluene disproportionation. The global world benzene production capacity will rise from estimated 41.8 Mt/y in 2009 to 55.8 Mt/y in 2015: http://marketpublishers.com/lists/5873/news.html, http://mcgroup.co.uk/researches/B/028500/Benzene.html. In 2005, the French production capacity was estimated at one Mt/year on seven industrial sites, i.e. mean of 143,000 t/year per site (www.ineris.fr/rsde/fiches/fiche_benzene_2005.pdf).

The Hydrodealkylation (Douglas, 1988) process for producing benzene, a classical benchmark in chemical process synthesis studies, is used in this paper (HDA process). This process involves two reactions: the conversion of toluene to benzene (Eq. (16)) and the equilibrium between benzene and diphenyl (Eq. (17)).

$$\text{Toluene} + H_2 \rightarrow \text{Benzene} + CH_4 \quad (16)$$

$$2\text{Benzene} \leftrightarrow \text{Diphenyl} + H_2 \quad (17)$$

Douglas (1988) has first extensively studied this process by using a hierarchical design/synthesis approach. The hydrogen feed stream has a purity of 95% and involves 5% of methane; this stream is mixed with a fresh inlet stream of toluene, recycled toluene, and recycled hydrogen. The feed mixture is heated in a furnace before being fed to an adiabatic reactor. The reactor effluent contains unreacted hydrogen and toluene, benzene (the desired product), biphenyl, and methane; it is quenched and subsequently cooled in a high-pressure flash separator to condense the aromatics from the non-condensable hydrogen and methane. The vapour steam from the high-pressure flash unit contains hydrogen and methane that is recycled. The liquid stream contains traces of hydrogen and methane that are separated from the aromatics in a low-pressure flash drum. The liquid stream from the low-pressure flash drum consisting of benzene, biphenyl and toluene is separated in two distillation columns. The first column separates the product, benzene, from biphenyl and toluene, while the second one separates the biphenyl from toluene, which is recycled back at the reactor entrance. Energy is saved by using the outlet stream leaving the reactor as its temperature is in the range of 620 °C, to preheat the feed stream coming from the mixer, via a heat exchanger (Fehe) some energy integration is achieved (Cao, Rossiter, Edwards, Knechtel, & Owens, 1998) (see Fig. 2).

As abovementioned, ARIANE^{TM} is used here both to compute the primary energy requirements of the process and to quantify the pollutant emissions related to energy production. A typical steam generating facility is considered: steam is produced from a conventional mono-fuel fired boiler (40 bar and 400 °C) and let down to lower pressure levels (respectively, 10 bar, 336 °C and 5 bar, 268 °C) through turbines which produce electricity used in the plant. ARIANE^{TM} is also used to model the fired heater (furnace) of the process as a bi-fuel fired heater (mix of natural gas and fuel).

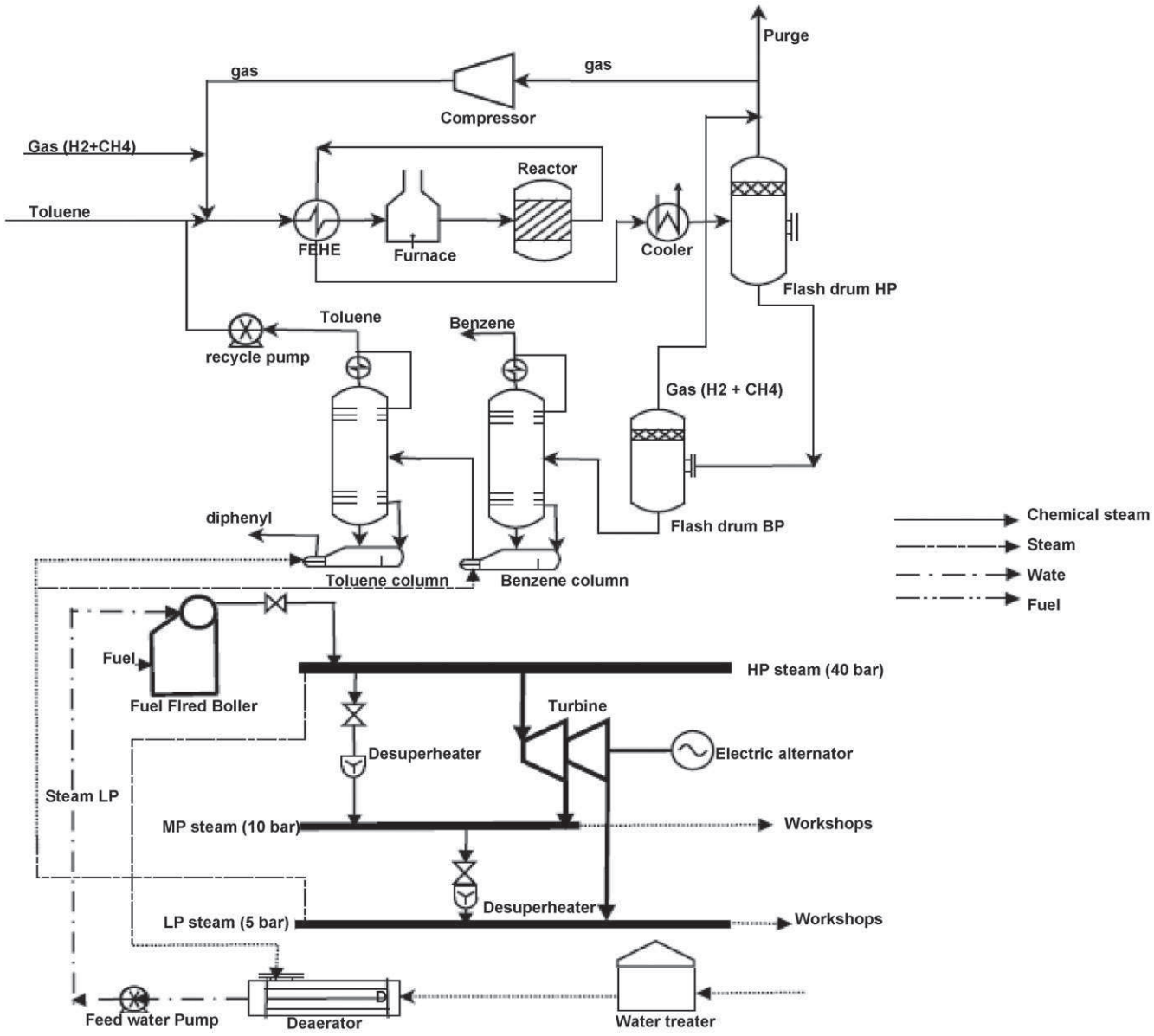

Fig. 2. HDA plant with is utility production unit.

To model the HDA process, commercial design and flowsheeting packages could be used in order to compute the objective functions. However, rather than using such packages, the equations proposed by Douglas (1988) have been directly implemented and solved by the Excel® solver. Biphenyl has been considered as a pollutant, and the environmental impact contributions for the components in the HDA process have been taken from Sikdar and El-Halwagi (2001).

4.2. Interaction between HDA process and utility production system

From the studies of Douglas (1988) and Turton, Bailie, Whiting, & Shaeiwitz (1997, 2009), seven variables were selected for HDA process because of their influence on the economic and environmental optimization criteria. These variables are initially used to compute the overall material balance of the main chemical compounds and as well as the associated thermodynamic properties (enthalpy, density, heat capacity, etc.) at various process nodes, with use of Simulis® Thermodynamics as a calculation server of thermodynamic properties. They serve finally to compute the size of all equipment items involved in the process for carrying out the required unit operations. Fig. 3 shows the interaction between the variables of the HDA process and the utility production system. HDA process operation needs thermal and electrical utilities. The thermal demand is formulated in terms of fuel flow rate to meet the need of heating the mixture in the furnace as well as of steam to meet the heating demands for the exchangers, reboiler and other items of the process. Eqs. (18)–(20) describe the demand of steam in the case of a reboiler. These requirements obtained from the energy balances of the HDA simulator are transmitted via Plessala™ as variables to the unit operations involved, which are modelled by use of Ariane™ (boiler, furnace). Then, fuel oil flow rate of, the natural gas flow rate and steam flow rate become secondary variables for Ariane™. The use of Ariane™ provides typical results related to the thermal power plant i.e. the power produced by the turbine and the flow rates of all the pollutants resulting from fuel combustion.

$$\dot{m}_{stmi} = \frac{Q_{bi}}{\Delta H_{stmi}} \tag{18}$$

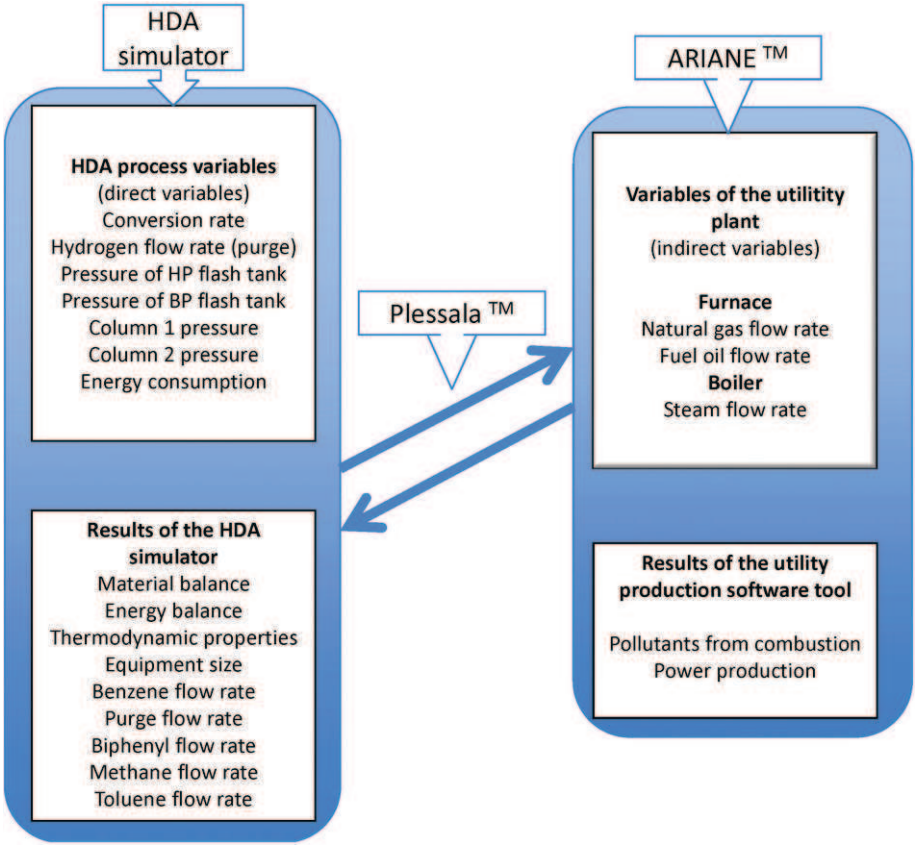

Fig. 3. Interaction between variables and simulation tools.

with $Q_{bi} = D_i h_{Di} + W_i h_{Wi} - F_i h_{Fi} + Q_{ci}$ (19)

and $Q_{ci} = D_i(R+1)\Delta H_{stri}$ (20)

4.2.1. Furnace modelling

The furnace is modelled as a bi-fuel boiler fed with both natural gas and fuel oil. Their flow rates are linked by the so-called energetic ratio and the furnace energetic demand as follows:

Consumed natural gas flow rate in the furnace: Q_{NG}

$$\dot{m} = \frac{Q_{Furnace}}{\eta \cdot LHV_{NG} \cdot ratio} \quad (21)$$

Consumed fuel oil flow rate in the furnace: Q_{FO}

$$\dot{m} = \frac{Q_{Furnace}}{\eta \cdot LHV_{FO} \cdot (1 - ratio)} \quad (22)$$

Energetic ratio:

$$ratio = \frac{\text{Energy provided by natural gas}}{\text{Total energy of natural gas and fuel oil}} \quad (23)$$

The fuels considered are natural gas (fuel 1) and fuel oil (fuel 2).

Case of a mono-fuel boiler:

The boiler is used to produce superheated vapour, for the operation of the turbine and hot water for the other units

$$\dot{m} = \frac{Q_{Boiler}}{\eta \cdot LHV_{NG}}. \quad (24)$$

4.2.2. Side stream turbine modelling

A boiler is used to product steam at high level of pressure and temperature, and then this steam is expended through a back-pressure and condensing turbine to generate power. The turbine is modelled with the following equations, implemented in ARIANE^{TM}:

$$S_i(T_i, P_i) = S_o(T_o^*, P_o) \quad (25)$$

Turbine maximum work

$$W_{\max} = H_i(T_i, P_i) - H_o(T_o^*, P_o) \quad (26)$$

Turbine isentropic yield

$$\eta = \frac{W_{\text{eff}}}{W_{\max}} \quad (27)$$

5. Optimization problem formulation

5.1. Economic objectives

As mentioned by Cutler and Perry (1983), a major objective of any industrial process is profit maximization. In this study, the benzene production (*ProdB*) to be maximized and the annual cost (*Annual cost*) have been chosen as economic functions. The benzene production is a direct output of the HDA process, while the annual cost is computed according to relations (28)–(31).

$$Annual \quad cost = 0.1\,\text{FCI} + C_{RM} + C_{UT} \quad (28)$$

FCI: Fixed capital investment ($)
0.1FCI: depreciation cost ($/y)
C_{RM}: cost of raw materials ($/y)
C_{UT}: cost of utilities ($/y)

$$\text{FCI} = \sum_i (Purchase \quad cost_i + Installed\, cost_i) \quad (29)$$

$$C_{RM} = \sum_i \dot{m}_{RMi} P_{RMi} \quad (30)$$

Table 1
Capital cost estimation for main items (*M&S*: Marshall & Swift Equipments Cost Index = 1, 468.6 (2009)) from Chemical Engineering January 2010.

Equipment investment cost ($)	Nonlinear form
Column cost	
D: column diameter (m)	Purchase cost $= 9.201\left(\frac{M\&S}{280}\right)(101.9D^{1.066}H^{0.802}F_c)$
H: column height (m)	
F_c: material pressure	Installed cost $= 9.202\left(\frac{M\&S}{280}\right)D^{1.066}H^{0.802}(2.18+F_c)+20.69\left(\frac{M\&S}{280}\right)4.7D^{1.55}HF'_c$
F'_c: material, tray space, tray type	
Exchanger cost	Purchase cost $= \left(\frac{M\&S}{280}\right)(474.7A^{0.65}F_c)$
A: heat exchanger area (m^2)	Installed cost $= \left(\frac{M\&S}{280}\right)(474.7A^{0.65})(2.29+F_c)$
Furnace cost	Purchase cost $= \left(\frac{M\&S}{280}\right)(5.52\times10^3)Q^{0.85}F_c$
Q: furnace absorbed (293 kW)	Installed cost $= \left(\frac{M\&S}{280}\right)(5.52\times10^3)Q^{0.85}(1.27+F_c)$

Table 2
Capital cost for the utility system.

Equipments investment cost (1000$)	Nonlinear form
Field erected boiler	$8.09F^{0.82}f_{p1}$
F: steam flow rate (kg/s);P: pressure (bar)	With, $f_{p1}=0.6939+0.01241P-3.7984\,\mathrm{Exp}(-3P^2)$
Steam turbine	
W_{st}: power (MW)	$25.79W_{st}^{0.41}$
Deaerator	
F: BFW flow rate (kg/s)	$0.41F^{0.62}$
Condenser	
Q: heat dissipated (MW)	$4.76Q^{0.68}$

$$C_{UT}=\sum_i \dot{m}_{UTi}P_{UTi} \tag{31}$$

$\dot{m}_{RMi}$: mass flow rate of raw material i (kg/h)
P_{RMi}: unit price of raw material i ($/kg)
$\dot{m}_{UTi}$: flow rate of utility i (kg/h, std m^3/h, m^3/h or kW)
P_{UTi}: unit price of raw material i ($/kg, $/std m^3, $/$m^3$ or $/kWh)

For the main equipment items, the purchase and installed costs were computed from the classical Guthrie's correlations (1969) (see Table 1). For the utility system, capital cost estimation was performed by using expressions developed in Bruno, Fernandez, Castells, and Grossmann (1998) (see Table 2). Finally, the costs of raw materials and utilities from Turton et al. (2009) are displayed in Table 3. For convenience, the *Annual cost* will be expressed in M$/y in what follows.

5.2. Environmental objectives

The environmental impacts are quantified by the so-called environmental burdens listed by IChemE. From the classification proposed by Azapagic, Emsley, and Hamerton (2003), eight categories of impacts can be identified, and environmental burden EB_i is computed as:

$$EB_i=\sum_{j=1}^{n} ec_i^j B_j \tag{32}$$

where ec_i^j is the estimated classification of the jth participant in the ith burden and B_j is the emission of the jth greenhouse gas. For a given application, the choice of particular impacts can be deduced from the flowsheet analysis, and they are quantified through mass and energy balances. Among the environmental impacts proposed by Azapagic et al. (2003), five representative items for the considered chemical process have been retained for composing the Environmental Burdens (EB_i). Each considered Environmental Burden can be briefly described as follows (see the study of Azapagic et al. (2003) for more information).

- Global Warming Potential (GWP in t CO_2 equivalent/y) which is a measure of how a given mass of greenhouse gas is estimated to contribute to the global warming. It is a relative scale, which compares the mass of a considered gas to the same mass of carbon dioxide. It is equal to the sum of eight emissions B_j of greenhouse gases multiplied by their respective GWP factors ec^j_{GWP}.
- Acidification Potential (AP in t SO_2 equivalent/y) is based on the contributions of SO_2 and NO_x to the potential acid deposition, that is on their potential to for H^+ protons. The five AP factors ec^j_{AP} are expressed relatively to the AP of SO_2.
- Photochemical Ozone Creation Potential (POCP in t C_2H_4 equivalent/y) (or PCOP) very often defined as summer smog, is the result of reactions that take place between nitrogen oxides NO_x and VOCs exposed to UV radiations. The POCP is related to ethylene as reference substance, that is to say that the POCP factors ec^j_{AP} for the eight concerned products are expressed relatively to the POCP of C_2H_4.
- Human Toxicity Potential (HTP in t C_6H_6-equivalent/y) expresses the potential harm of chemicals released into the environment. HTP includes both inherent toxicity and generic source-to-dose relationships for pollutant emissions. It uses a margin-to-exposure ratio to evaluate the potential for health impact from

Table 3
Cost of raw materials and utilities used in HDA process.

Raw materials	Cost ($/kg)
Toluene	0.648
Hydrogen	1
Utilities	Cost ($/common unit)
Fuel oil (n°2)	$549/$m^3$
Natural gas	$0.42/std m^3
Electricity	$0.06/kWh
Steam from Boiler	
High pressure	$29.97/1000 kg
Medium pressure	$28.31/1000 kg
Low pressure	$27.70/1000 kg
Cooling tower water (30–40 °C)	$14.8/1000 m^3

exposure to harmful agents, both carcinogens and non carcinogens. It is calculated by adding human toxic releases to three different media, i.e. air, water and soil. The toxicological factors are calculated using the acceptable daily intake or the tolerable daily intake of the toxic substances. The human toxicological factors (26 from Azapagic et al., 2003) are still at development stage, so that HTP can only be taken as an indication and not as an absolute measure of the toxicity potential.

- Eutrophication Potential (EP in t PO_4^{3-} equivalent/y) is defined as the potential of nutrients to cause over-fertilisation of water and soil which in turn can result in increased growth of biomass.

It must be emphasized at this level that the approach presented here does not account for the entire life cycle of the process since the environmental burdens are associated with the emissions generated by the process both from the material components and from the energy vectors. For instance, the outcome of benzene is not considered here as it is viewed as the desired product for the process. We are thus aware that the approach developed here does not embed the whole life cycle of the product. Moreover, the extraction resource associated with raw materials such as toluene is not included. Briefly, only the process contribution is considered in this study.

Of course, a life cycle assessment methodology could be attractive to be more general and the abovementioned drawbacks could be partially overcome by using standard environmental databases available in the literature (e.g. EcoInvent for instance http://www.ecoinvent.ch/) implemented in life cycle assessment software tools. This issue will constitute a perspective of this work.

5.3. Multiobjective problem formulation

Using the previous economic and environmental objective functions defined above, the following multiobjective nonlinear optimization problem is formulated as follows: determine decision variables (process operating conditions) in order to:

$$\text{Max}\,(ProdB) \tag{33}$$

$$\text{Min}\,(Annual\ cost) \tag{34}$$

$$\text{Min}\,(EB_i), \quad i = 1, \quad 5 \tag{35}$$

s.t.

Mass and energy balances (Excel® and ARIANE™)

Bounds on decision variables

ProdB represents the benzene production at the top of column 2 (in kmol/h).

The additional following constraints have been considered in the Multigen interface, the numerical values value have been deduced from the analysis of Douglas (1988) and Turton et al. (1997).

- The purity of the benzene product is at least 99.97%
- The hydrogen feed must have a purity of 95%
- The reactor outlet temperature is less than 704.50 °C
- The quencher outlet temperature is less than 621.16 °C
- The conversion rate C is bounded as: $0.5 \leq C \leq 0.9$
- The hydrogen flow rate purged F_{PH} (kmol/h) is bounded as: $30 \leq F_{PH} \leq 300$
- All pollutants, CO_2, NO_x, CO, SO_2, dusts flow rate (kg/h) must take only positive values.

5.4. GA parameters

The two classes of optimization problems presented below have been solved by using the code NSGA IIb of the Multigen toolbox, with the following operating parameters: population size = 200, number of generations = 200, Crossover rate = 0.75 and Mutation rate = 0.2.

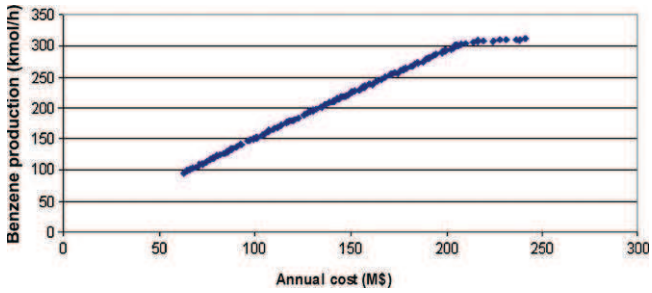

Fig. 4. Benzene production vs. *Annual cost*.

Douglas (1988) then Turton et al. (1997) define bounds on the HDA variables as reported in Table 4. The initial population was randomly generated between these bounds, and a particular set of variables has been extracted from the initial population (column Initial value of Table 4) for further comparison purposes (see Table 5 line "Initial").

6. Case 1: economic followed by ecological study

6.1. Solution strategy

For the problem involving all the objectives, the Pareto front is a cloud of points located in a hypercube of $\Re^7$. To reduce this hypercube dimension in order to interpret as good as possible the genetic algorithm results, a two steps solution strategy is implemented. In the first step, the biobjective problem ((33) and (34)) is solved to obtain an efficient trade-off (*ProdB**, *Annual cost**) between the maximal benzene production and the minimal investment cost. Then the benzene production, being fixed at a value *ProdB** obtained from the Pareto curve, the problem ((33)–(35)) is solved for the investment cost lying in a low range [*Annual cost**, *Annual cost** + α], with a value of α equal to 15% for instance. A degradation of the annual cost is also allowed, thus improving the environmental performances of the process of benzene production.

6.2. Economic study: biobjective optimization (ProdB, Annual cost)

The Pareto front corresponding to the optimization of the pair (*ProdB*, *Annual cost*) is displayed in Fig. 4. The curve is composed of two portions of straight lines. The knee located on the curve gives the best trade-off (Branke et al., 2004), that is to say (205.04 M$, 299.96 kmol/h). Without environmental considerations, this solution would be adopted for plant design. In the following, the benzene production will be lower bounded at 300 kmol/h and the annual cost will vary in the range [205.04 M$, 299.96 M$] in order to compete with additional environmental objectives. The values of all considered objectives for the first point (204.5, 299.9) noted "First" of the flat line of Fig. 4 and the initial point described in Table 4 are used for comparing objectives in Table 5.

6.3. Study of environmental burdens via biobjective optimizations

The benzene production being fixed at 299.96 kmol/h and the *Annual cost* lying in the range [205.04 M$, 299.96 M$], the criteria are optimized by pairs for identifying redundant objectives expressed in terms of multilinear functions of the three remaining ones (*Annual cost*, EP and AP). These dependent impacts (GWP, HTP and POCP) will be removed from the multiobjective optimization problem, where some degradations of the *Annual cost* are allowed

Table 4
Decision variables for the HDA process.

Decision variables	Lower bound	Initial value	Upper bound
Conversion rate (%)	0.5	0.75	0.9
Hydrogen purge flow rate (kmol/h)	31	198	308
Flash pressure (bar)	30	34	34
Stabilizer pressure (bar)	4	10	10
Column 1 pressure (bar)	2	2	4
Column 2 pressure (bar)	1	1	2
Ratio (bi-fuel furnace) (%)	0.1	0.85	0.9

Table 5
Values of objectives.

Objective	*ProdB* t/y	*Annual cost* M$/y	EP t PO_4^{3-}/y	AP t C_2H_4/y	GWP t CO_2/y	HTP t C_6H_6/y	POCP t C_2H_4/y
Initial	305.00	277.42	9759.06	11,190.34	1,884,528.48	18,699.58	2472.32
First	299.96	205.04	13,831.12	4829.26	1,410,428.13	26,537.88	1944.11
Gain vs. initial (%)	−1.62	26.00	−29.44	56.85	25.16	−41.91	21.36

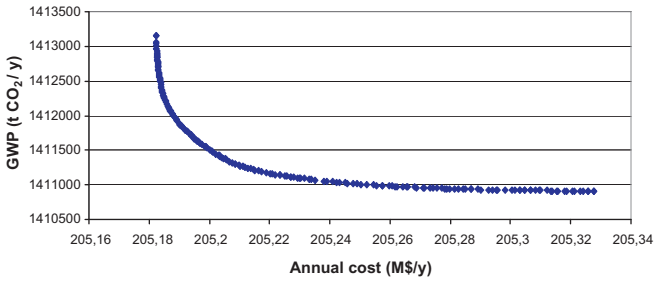

Fig. 5. GWP vs. *Annual cost*.

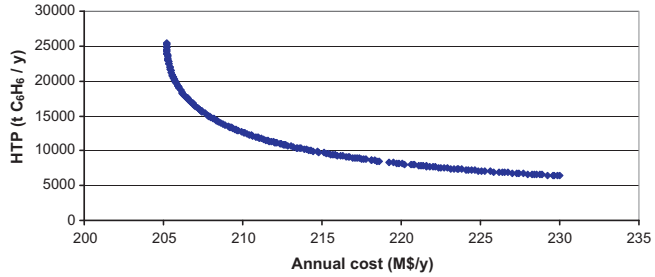

Fig. 6. HTP vs. *Annual cost*.

for globally improving all the environmental impacts (including the redundant ones).

(GWP, *Annual cost*): Non dominated points appear only in low ranges for the two objectives (see Fig. 5), suggesting a strong relation between them (see section 5.4). **(HTP, *Annual cost*)**: The Pareto front is displayed in Fig. 6, where the two objectives have opposite effects. **(EP, *Annual cost*)**: The results are shown in Fig. 7. The curves displayed in Figs. 6 and 7 exhibit very similar trends, due to a strong link between them (see Section 5.4). **(AP, *Annual cost*):** The Pareto

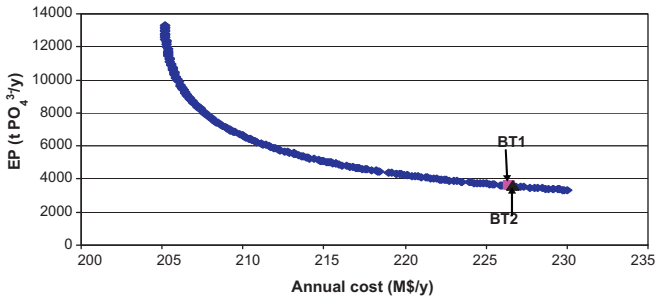

Fig. 7. EP vs. *Annual cost*.

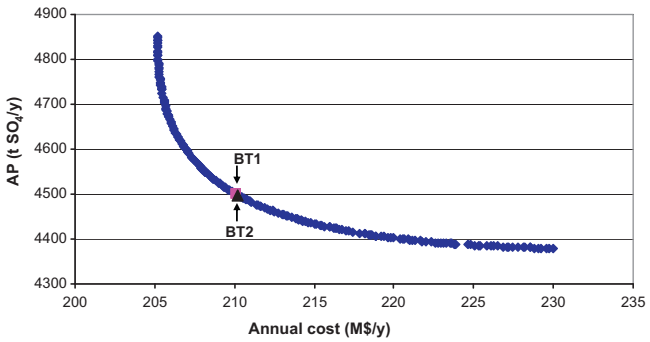

Fig. 8. AP vs. *Annual cost*.

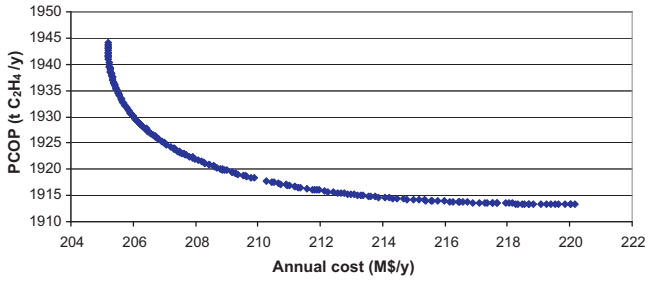

Fig. 9. POCP vs. *Annual cost*.

front is displayed in Fig. 8. **(POCP, *Annual cost*)**: the results reported in Fig. 9 show that non dominated points appear only in a low range for the *POCP* objective, suggesting a strong relation between these two functions. **(AP, EP)**: concerning the pair (AP, EP), the biobjective optimization gives the front displayed in Fig. 10, where it can be pointed out that the two impacts exhibit antagonist behaviours.

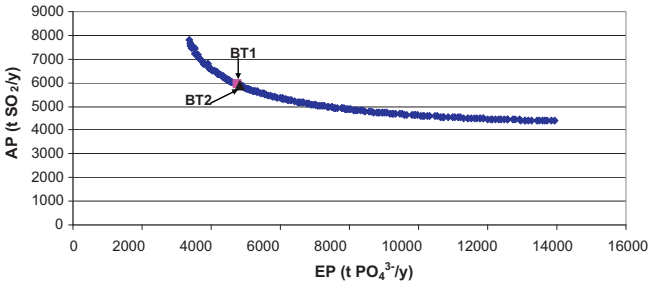

Fig. 10. AP vs. EP.

Table 6
Results of the multilinear regressions – Case 1.

Objective	*Annual cost* M$/y	EP t PO_4^{3-}/y	AP t C_2H_4/y	*y*-Intercept	Coeff. corr.	Error max (%)
GWP t CO_2/y	5445.29	−0.64	43.95	90,529.67	0.9988	0.51
HTP t C_6H_6/y	-6.29×10^{-5}	1.92	-3.70×10^{-5}	9.25×10^{-3}	1.000	10^{-6}
POCP t C_2H_4/y	0.44	4.07×10^{-3}	8.67×10^{-2}	1377.95	0.9999	0.03

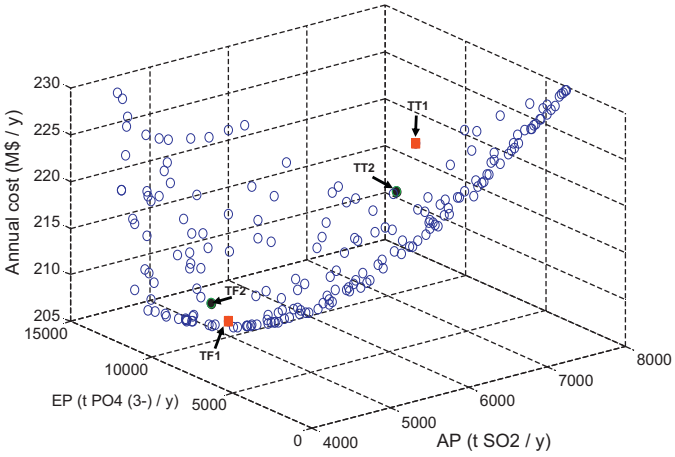

Fig. 11. Triobjective optimization (*Annual cost*, EP, AP) – Case 1.

6.4. Redundant objectives

From the previous section, environmental impacts GWP, HTP and POCP were found to be linked with other objectives *Annual cost*, EP and AP. From 200 randomly generated values for the independent variables (defined in Table 4), the objectives *Annual cost*, EP, AP, GWP, HTP and POCP were computed and multilinear regressions were performed on the Excel platform. The experiment was carried out several times and gives the following results of Table 6.

In all cases, the coefficient of correlation is very good. From the coefficients of multilinear equations, it can be observed that GWP and POCP are mainly increasing functions of the *Annual cost*, while HTP depends principally on EP. So impacts GWP, POCP and HTP being explicit functions of terms *Annual cost*, EP and AP, they can be suppressed from the following multiobjective optimization phase.

6.5. Triobjective optimization

From the previous biobjective study, only three independent objectives are remaining: *Annual cost*, EP and AP. They are now simultaneously optimized, and the results are displayed in Fig. 11. The flat portion of the cloud of points near *Annual cost* ≈ 205 M$/y, EP ≈ 10,000 t PO_4^{3-} and AP ≈ 5000 t SO_2/y suggests that good solutions may exist in this zone for the three objectives.

Table 7
Results of TOPSIS analysis for biobjective optimizations.

Pair	*Annual cost* M$/y	EP t PO_4^{3-}/y	AP t SO_2/y
(EP, Cost) 1	226.42	3590.12	
(EP, Cost) 2	226.64	3574.40	
(AP, Cost) 1	210.13		4499.36
(AP, Cost) 2	210.18		4498.29
(AP, EP) 1		5918.37	4763.31
(AP, EP) 2		5886.94	4819.97

6.6. Selecting solutions

6.6.1. Biobjective optimizations

From the three independent objectives, *Annual cost*, EP and AP, a TOPSIS analysis is carried out twice for the three possible pairs in order to detect in each case the two best points called BT1 and BT2 (BT signify Biobjective Topsis) in Figs. 7, 8 and 10. The results are indicated in Table 7. Note that the FUCA procedure cannot be implemented on biobjective problems, insofar as the Pareto front being per definition a set of non dominated points, the sum of ranks is constant and equal to the number of points in the front plus one (the best point for one objective being the worst one for the other).

6.6.2. Triobjective optimization

A TOPSIS and a FUGA analysis are carried out twice on the global set of objectives (*Annual cost*, EP, AP, GWP, HTP and POCP). In these studies even the dependant objectives GWP, HTP and POCP, computed by using Table 6 from the three independent ones, have to be considered, because they can influence the decision making. However, the results are only displayed for the three independent criteria in Fig. 11. Let us point out that the rankings were performed without any preference factor. The two best solutions obtained from TOPSIS (respectively FUCA) are called TT1 and TT2 (respectively TF1 and TF2) on the 3D curve.

Table 8 exhibits the results obtained both for the three independent objectives and for the three dependent ones in the last columns. For each objective the gain in % is computed vs. solution called “first” in Table 5. The values of the corresponding variables are given in Table 9. Obviously, the solutions obtained when projecting 3-D solutions in the corresponding 2-D spaces globally over estimate the solutions obtained in the pure 2-D optimizations. As a partial conclusion, preliminary 2-D optimizations cannot be used for estimating the solutions of the 3-D problem. The TOPSIS and FUCA rankings provide yet surprisingly with scattered values. The FUCA procedure gives results that are much more in agreement

Table 8
Results of TOPSIS and FUCA analysis for the triobjective optimization.

Case	*Annual cost* M$/y	EP t PO_4^{3-}/y	AP t SO_2/y	GWP t CO_2/y	HTP t C_6H_6/y	POCP t C_2H_4/y
TT1	227.34	4404.41	6221.15	1,599,077.01	8439.0316	2038.64
Gain	−10.88	67.08	−28.82	−13.36	68.20	−4.86
TT2	222.28	4735.06	6043.41	1,563,468.01	9072.59	2022.27
Gain	−8.41	64.61	−25.14	−10.85	65.81	−4.02
TF1	206.99	9770.38	4930.16	1,428,118.43	18,720.83	1939.25
Gain	−0.95	26.98	−2.09	−1.25	29.46	0.25
TF2	209.03	10,109.41	4782.53	1,432,472.39	19,370.44	1928.70
Gain	−1.95	24.45	0.97	−1.56	27.01	0.79

Table 9
Values of the decision variables for the triobjective optimization.

Decision variables	TT1	TT2	TF1	TF2
Conversion rate of toluene	0.59	0.60	0.70	0.75
Hydrogen purge flow rate (kmol/h)	300	300	300	300
Flash pressure (bar)	33.98	33.98	33.98	33.88
Stabilizer pressure (bar)	9.99	9.71	10	9.97
Column 1 pressure (bar)	3	3	3	3
Column 2 pressure (bar)	1.68	1.21	1.31	1.02
Ratio (bi-fuel furnace)	0.29	0.32	0.24	0.34

with a simple graphical analysis: besides, the global gain in the set of objectives is higher than the value computed by the TOPSIS analysis. A closer examination of TF1 and TF2 finally leads to select the solution TF2, because it gives only one negative gain in environmental impacts. In the following studied case, only the FUCA ranking will be used.

7. Case 2: simultaneous economic and ecological studies

7.1. Context

Let us consider now that according to the EPA's more stringent regulations, the GWP must be below a threshold of 1,400,000 t CO_2/y, which may have some effects on production level. Besides, incentive measures for environment protection lead to improve simultaneously the other environmental objectives EP, AP, HTP and POCP (as compared with the previous case).

After randomly generating several thousands of values for independent variables, it appears that values of GWP slightly lower than 1,400,000 t CO_2/y are located in a benzene production range of [250,280] kmol/h. So the multiobjective optimization is now carried out in this range of production. The same strategy as in the previous case consisting in decoupling objectives in dependent and independent sets is implemented again. In order to avoid extrapolation model problems that may occur, multilinear regressions are performed again for dependent objectives. Finally, a triobjective optimization is carried out for identifying the best solution under these new environmental constraints.

7.2. Multilinear regressions

For 200 values of independent variables within a production range [250,280] kmol/h randomly generated, the values of the other objectives are computed and the results of multilinear regressions are displayed in Table 10.

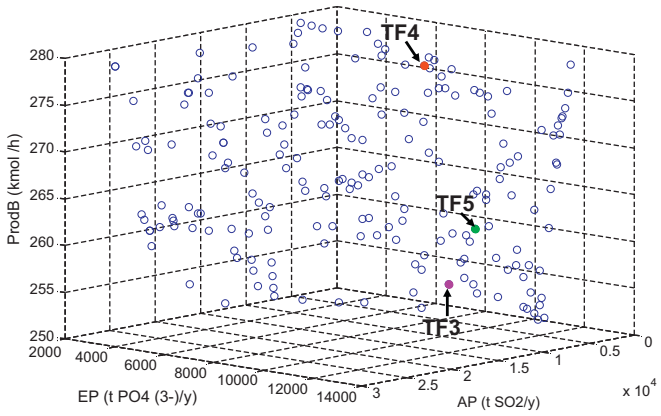

Fig. 12. Triobjective optimization (*ProdB*, EP, AP) – Case 2.

7.3. Triobjective optimization

A triobjective optimization is carried out on the three independent objectives (*ProdB*, EP and AP), the other dependant objectives *GWP*, *HTP*, *POCP* and *Annual cost*, being computed by using Table 10 from the three independent ones. The Pareto curve obtained from the tricriteria optimization of (*ProdB*, EP, AP) is displayed in Fig. 12, where the points called TF3, TF4 and TF5 correspond respectively to the best solution found by performing a non weighted (resp. weighted) sum of ranks of the seven objectives.

7.4. Selecting solutions

A FUCA ranking is first carried out with the same weight for all the objectives. The best solution noted TF3 is identified among the five first ones. However, in this situation, benzene production is improved only by 14% .The goal of any industrial process being firstly profit maximization, a second FUCA ranking is performed with different weights on objectives: 0.4 for benzene production

Table 10
Results of the multilinear regressions – scenario 2.

Objective	*ProdB* kmol/h	EP t PO_4^{3-}/y	AP t C_2H_4/y	*y*-Intercept	Coeff. corr.	Error max (%)
GWP t CO_2/y	4901.44	−11.61	23.36	10,945.95	0.9397	3.75
HTP t C_6H_6/y	-2.14×10^{-5}	1.92	-3.68×10^{-5}	1.66×10^{-4}	1.000	6×10^{-6}
POCP t C_2H_4/y	5.08	2.04×10^{-3}	7.950×10^{-2}	5.46	0.9942	0.69
COST M$/y	0.6959	0	0	−5.03502	0.9997	2.10

Table 11
Results of FUCA analysis for the triobjective optimization.

Case	Benzene Prod (kmol/y)	*Annual cost* M$/y	EP t PO_4^{3-}/y	AP t SO_2/y	GWP t CO_2/y	HTP t C_6H_6/y	POCP t C_2H_4/y
TF3	253.50	171.30	7901.62	3737.16	1,249,024.11	15,104.58	1599.81
Gain	−16.34	19.08	21.84	21.86	12.81	22.02	17.05
TF4	276.73	188.21	7149.41	4446.35	1,388,198.24	13,666.63	1772.36
Gain	−9.49	18.04	29.28	7.03	3.09	29.45	8.11
TF5	259.88	175.95	8982.89	3799.92	1,269,260.85	17,171.37	1639.400
Gain	−14.23	15.82	11.15	21.86	11.39	11.35	15.00

Table 12
Values of the decision variables for the triobjective optimization – Case 2.

Decision variables	TF3	TF4	TF5
Conversion rate of toluene	0.74	0.71	0.76
Hydrogen purge flow rate (kmol/h)	299.30	299.92	299.92
Flash pressure (bar)	29.27	28.73	32.57
Stabilizer pressure (bar)	8.60	6.02	7.00
Column 1 pressure (bar)	2.13	2.03	2.21
Column 2 pressure (bar)	1.02	1.33	1.74
Ratio (bi-fuel furnace)	0.16	0.24	0.17

and 0.1 for the six criteria. As in the previous case, the best solution noted TF4 is identified among the five first ones. Finally, a compromise solution TF5 generated with the respective weights of 0.35 for the benzene production, 0.15 for the GWP and 0.1 for all the others is also determined.

Table 11 exhibits the results obtained both for the three independent objectives and for the three dependent ones. For each objective the gain in % is computed vs. solution TF2 of Table 8. The values of the corresponding variables are given in Table 12.

8. Conclusions

This paper has presented a methodology for eco-design and optimization of a chemical process taking into account the contribution of utility generation, via the industrial software ARIANE^{TM}. The well-known benchmark HDA process first developed by Douglas (1988) illustrates the approach, which is totally different with the traditional end-of-pipe treatment methods. The process was designed under classical engineering objectives like benzene production and total annual cost, by also considering classical environmental burdens as the Global Warming Potential, the Acidification Potential, the Photochemical Ozone Creation Potential, the Human Toxicity Potential and the Eutrophication Potential. A variant of the classical multiobjective genetic algorithm NSGA II was used for solving the various multiobjectives problems.

In a preliminary study, a good value of the benzene production was identified on a limited range of costs, then a possible degradation of the annual cost in this range, and thus of the benzene production were allowed to improve the environmental performances of the process. Instead of searching for a Pareto front on the whole set of objectives, a preliminary study of objectives was carried out for determining a sub-set of dependent criteria expressed as multilinear functions of the remaining independent ones. So the multiobjective problem was reduced to a tricriteria one. The values of corresponding dependent objectives were computed from the independent ones, and a TOPSIS and FUCA analyses were performed on the whole set of objectives, FUCA giving much better results than TOPSIS. An alternative would be to identify an ideal point on the Pareto front by defining a metric distance to an utopia point (i.e. minimizing all objectives for instance) as an objective function using a single objective GA.

A second scenario is based on another formulation for which a more stringent environmental regulation has to be satisfied, expressed as the GWP criterion to be lower than a given threshold. The objective also has positive gains over all environmental objectives EP, AP, HTP and POCP. Multilinear regressions were performed again, to obtain a new tricriteria problem, solved again by means of NSGA II. By simply tuning the weighting factors used in the FUCA ranking, several solutions offering a good trade-off can be deduced for objectives. Here three solutions corresponding respectively to high, medium and low ranges of benzene production are generated. This methodology can be applied to a wide spectrum of chemical process design problems with multiple and environmental objectives and makes explicit the trade-offs between them.

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