A NEW BENCHMARK FOR PLANTWIDE PROCESS CONTROL

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Abstract - The hydrodealkylation process of toluene (HDA) has been used as a case study in a large number of control studies. However, in terms of industrial application, this process has become obsolete and is nowadays superseded by new technologies capable of processing heavy aromatic compounds, which increase the added value of the raw materials, such as the process of transalkylation and disproportionation of toluene (TADP). TADP also presents more complex feed and product streams and challenging operational characteristics both in the reactor and separator sections than in HDA. This work is aimed at proposing the TADP process as a new benchmark for plantwide control studies in lieu of the HAD process. For this purpose, a nonlinear dynamic rigorous model for the TADP process was developed using Aspen Plus™ and Aspen Dynamics™ and industrial conditions. Plantwide control structures (oriented to control and to the process) were adapted and applied for the first time for this process. The results show that, even though both strategies are similar in terms of control performance, the optimization of economic factors must still be sought.
Keywords: Plantwide control; Aromatic complex; TADP process; Dynamic simulation.

INTRODUCTION

The use of recycles and heat integration in the transformation processes is a consolidated solution to increase yields and to reduce operational costs. These factors tend to increase the process complexity, demanding a control perspective not limited to the analysis of the individual units.

Many authors point out that the need for a plantwide perspective on control arises mainly due to these changes in the way plants are designed. Indeed, these factors lead to more interactions and therefore the need for a perspective beyond individual units, as pointed out by Stephanopulos (1984) and earlier by Buckley (1964). Larsson & Skogestad (2000) clari-

fied that the term plantwide control does not mean the tuning and analysis of the behavior of each control loop, but rather the control philosophy of the overall plant with emphasis on structural decisions (Morari, 1982), such as selection of manipulated ("inputs"), controlled ("outputs") and measured variables ("extra-outputs"); design of control configuration (a structure interconnecting outputs, setpoints and manipulated variables) and selection of controller type. These decisions are all taken during the basic design conception, unfortunately before the complex control studies that in general are not performed by process engineers.

Myers (1997) defined the aromatic complex as a combination of process units that can be used to

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convert petroleum naphtha and pyrolysis gasoline (pygas) into the basic petrochemical intermediates: benzene, toluene, and xylenes know as BTX.

Benzene, toluene and xylenes are produced through the catalytic reforming of naphta, but the thermodynamic proportion obtained in this process (32:36:32) is different from the market demand (55:11:34). Toluene has the lower demand and several processes are used to convert toluene and to produce, preferentially, benzene and xylenes (Serra *et al.*, 2004).

This work focuses on the plantwide control of an aromatic complex, more precisely the Transalkylation and Disproportionation Unit (TADP), whose function is the increase of xylenes and benzene production in the aromatic complex from surplus toluene. This process is considered to be a technological improvement of the HDA process because, besides benzene, the TADP process also produces xylenes and generates fewer by-products with low commercial value.

For this purpose, a nonlinear dynamic rigorous model for the TADP process was developed using Aspen Plus™ and Aspen Dynamics™ and techniques previously employed in the HDA process were investigated and compared in order to create an effective control structure for the TADP process. To the knowledge of the authors, this is the first study of plantwide control applied to the TADP process.

PLANTWIDE CONTROL

According to Qiu *et al.* (2003), a major problem in controlling a plant is to develop effective control structures for the entire complex. For Larsson & Skogestad (2000), the design of a control structure is difficult to define mathematically, especially because of the size and cost involved for the precise formulation of the problem. This is the mathematically-oriented approach (or design of the control structure), i.e, the systematic approach for solving the plantwide control problem. An alternative is the development of heuristics based on experience and understanding of the process and is referred to as the process-oriented approach (Luyben, 2002; Luyben *et al.*, 1998).

The implementation of the methodology from Luyben *et al.* (1998), using the process-oriented approach, is composed of nine steps: i. establish control objectives; ii. determine the control degrees of freedom; iii. establish energy inventory control; iv. set production rate; v. define product quality and safety control; vi. define inventory control; vii. check component balances; viii. control unit operations individually and ix. use remaining control degrees of freedom to optimize economics or improve dynamic controllability.

Morari (1982) stated that "in search of a control structure considered optimizing, the main objective is to incorporate the economic objectives to the process control objectives". In other words, "the goal is to find a function $c(u,d)$ of process variables, that, when held constant, leads the manipulated variables automatically to their optimal working values, and with it, to the optimal operating conditions [...]." This means that, keeping $c(u,d)$ in their reference values c_s, through the manipulated variables u, and under several disturbances d, the process is operating at its optimal steady-state.

Larsson & Skogestad (2000) introduced the concept of a "self-optimizing" control system, which consists of determining the best set of controlled variables in a manner that results in an economic performance of the overall process closest to the optimal value of the economic objective function. They evaluated the effects of a loss function (departure from optimum) in the implementation of the reference value of the controlled variable.

The authors presented a design procedure based on a mathematically-oriented approach, but with some elements of the process-oriented approach. The procedure starts with a top-down analysis to select the controlled variables, based on ideas of self-optimization. At this stage, a rigorous steady-state model is needed and the operational objectives (economic steady-states) have to be defined. The result consists of one or more alternative sets of controlled variables.

This top-down analysis is followed by a bottom-up analysis, starting with the regulatory control layer. After this stage, the setpoints of the regulatory layer and some unused manipulated variables are the remaining degrees of freedom, which can be used to control the primary controlled variables. This control layer is called the supervisory layer. Two main approaches are possible for this layer: single-loops (decentralized) controllers with feedforward connections, or multivariable control. According to the authors, appropriately designed multivariable controllers will have better performance, but this must be negotiated against the cost of obtaining and maintaining the models used in the controllers. In the sequence, an optimization layer is applied with the purpose to identify active constraints and compute optimal set-points c_s for the controlled variables. Finally, nonlinear dynamic simulations should be performed to validate the proposed control structure.

As pointed out by Qiu *et al.* (2003), the HDA process has all the characteristics for plantwide control

application. Luyben *et al.* (1998) conducted a case study with the HDA unit applying their design procedure of plantwide control with nine steps. Araújo *et al.* (2007a) applied the concept of self-optimizing control from Larsson & Skogestad (2000) to show that several authors did not specify clearly all thirteen degrees of freedom present in the steady-state of the HDA process. The strategies of Luyben *et al.* (1998) and Araújo *et al.* (2007a) were adapted and compared in the present contribution to the TADP process. They were chosen after a literature review since they are up-to-date and representative of typical (process- and mathematically-oriented) plantwide control approaches.

DESCRIPTION OF THE TADP PROCESS

The technologies for transferring the methyl group convert toluene to benzene and xylenes at the same time, while the hydrodealkylation scheme mainly produces benzene. Due to the growing demand for xylenes, the process of transfer of the methyl group is preferred. Compared with the reactions of methyl group transfer, the process of hydrodealkylation operates at higher temperatures and re requires higher

operating costs and a capital investment about 10% higher (Jeanneret *et al.*, 2003).

The main products of the disproportionation of toluene are benzene and xylenes. There are also some side reactions including disproportionation of xylenes producing A10 (aromatic with 10 carbons) and dealkylation of alkylbenzenes producing light gases. A diagram of the generic TADP process is presented in Figure 1.

The feed containing toluene, A9/A10 and hydrogen (including both the recycle and make-up) passes through a bed of zeolite in the reactor. Then the products benzene, xylene and A10+ are separated at the overhead of the benzene column, xylene column and in the bottom of the heavy aromatics column, respectively. Toluene and A9/A10 are separated at the top of the toluene and heavy aromatics columns, respectively, and recycled to increase the conversion. The hydrogen is recycled from the first separator through a recycle compressor. Transalkylation reactions are controlled by thermodynamics, where the efficiency ratio in xylene and xylene/benzene increases with increasing content of A9+ in the feed. As the price of aromatic A9+ is usually lower than the BTX aromatic compounds, increasing the content of A9+ in the feed also reduces the cost of transalkylation.

Figure 1: Generic TADP Process**.**

PROCESS MODELING AND STEADY-STATE SIMULATION

Introduction

For the model development, the commercial simulator Aspen Plus™ v. 2006.5 was chosen. Figure 2 presents the flowsheet of the assumed process. Some changes in the generic TADP process were implemented aiming a closer approximation to the concept of a petrochemical complex with the inclusion of input streams from adjacent units. The main changes were: the inclusion of a feed stream of a mixture of benzene and toluene (bentol), from a hypothetical reprocessing complex; the xylene and heavy aromat-

ics columns were removed from the study (aiming an approximation with the HDA process) for comparison purposes; a fresh stream of toluene was added in the feed of the benzene column, since toluene is not a product of the complex and therefore this stream can present variations in compositions that are absorbed in the separation columns. Sidestreams were included in the stabilizer column and in the benzene column based on industrial plant design information of real units (Klafke, 2011).

The kinetic model is presented in the Appendix and was proposed by Ouguan *et al*. (2007). The design data were based on information of industrial units (Klafke, 2011) and are summarized in Table 1 and Table 2. The streams data are also available in Klafke (2011).
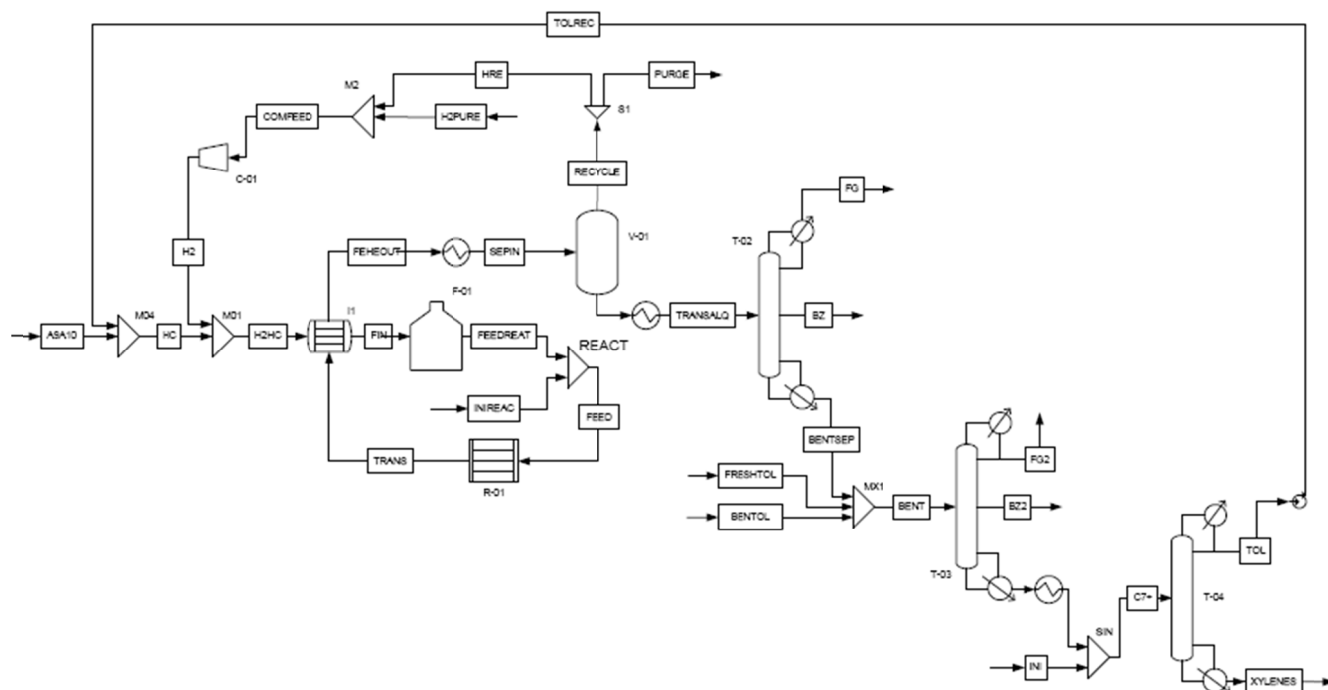
Figure 2: Process Flowsheet.

Table 1: Specification of the distillation columns.

	Distillation Columns		
	T-02	T-03	T-04
Number of stages	35	44	52
Pressure at the top [Pa g]	61.2	6.1	59.1
Feed stage	24	24	24
Sidestream stage	5	7	-
Diameter [m]	3.9	5.8	5.7
Specification 1 (mass basis)	Boilup ratio = 2	Distilled = 10^{-11} kg/h	Distilled = 144338 kg/h
Specification 2 (mass basis)	Ratio distilled/feed = 0.018	Boilup ratio = 5.748	Reflux ratio = 1.4
Dimensions of the reflux and reboiler – Diameter x Length (m)			
Reflux drum	1.91 x 5.7	3.7 x 11	2.8 x 8.3
Base drum	3.4 x 10	4.6 x 13.9	4 x 12

Table 2: Equipment dimensions and specifications.

Reactor	Length	8	m
	Diameter	5.3	m
FEHE (Feed Effluent Heat Exchanger)	UA	2823.5	kJ/(s°C)
Separator	Length	10	m
	Diameter	3	m

Steady-State Analysis for Plantwide Control

Degrees of Freedom Analysis

The number of steady-state degrees of freedom determines the number of steady-state controlled variables that can be chosen. In complex processes, it is useful to sum the number of degrees of freedom of the individual units, as given in Table 3. From these rules, the degrees of freedom are calculated according to Table 4. This analysis can be verified by a balance of the 22 manipulated variables considered in this process (see Table 5). However, 7 of the original degrees of freedom are flowrates used to stabilize liquid levels with no steady-state effect. Thus, there are 22–7 = 15 degrees of freedom, as shown in Table 4.

For this process 49 variables were considered as controlled variables. This selection is presented in Table 6. With 15 degrees of freedom and 49 candidates for controlled variables, an analysis of all possible structures is impractical. To avoid this combinatorial explosion, the active constraints are first determined and then an optimization analysis can be applied to define the remaining set.

Table 3: Typical number of steady-state degrees of freedom for process units, based on Araújo *et al*. (2007b).

Process Unit	Degrees of Freedom
Each external feed stream	1 (feedrate)
Splitter	n-1 split fractions (n is the number of exit streams)
Mixer	0
Compressor, turbine, and pump	1 (work)
Adiabatic flash tank	0^*
Liquid phase reactor	1 (holdup)
Gas phase reactor	0^*
Heat Exchanger	1 (duty or net area)
Columns (e.g. distillation) excluding heat exchangers	0^* + number of side streams

*Add 1 degree of freedom if pressure is set (need an extra valve, compressor, or pump).

Table 4: Number of steady-state degrees of freedom analysis.

Process Unit	Degrees of Freedom
External feed streams	$4 \times 1 = 4$
Splitters (purge)	$1 \times 1 = 1$
Compressor[*]	$1 \times 0 = 0$
Adiabatic flash[**] (separator)	$1 \times 0 = 0$
Gas phase reactor [**]	$1 \times 0 = 0$
Heat exchangers in recycle section [***] (furnace and cooler)	$2 \times 1 = 2$
Heat exchangers in three distillation columns	$3 \times 2 = 6$
Three distillation columns, two of it with one sidestream each	$0 + 2 \times 1 = 2$
Total	15

* Considering fixed power in the compressor.
** Assuming no adjustable valves for pressure control (fully open valve ahead of the separator).
*** The FEHE duty is not a degree of freedom because there is no adjustable bypass.

Table 5: List of manipulated variables.

	Manipulated Variable	State
U1	A_9/A_{10} feed flow rate	Steady State
U2	Fresh gas feed flow rate	Steady-state
U3	Fresh toluene flow rate	Steady-state
U4	Bentol flow rate	Steady-state
U5	Furnace heat duty	Steady-state
U6	Cooler heat duty	Steady-state
U7	Purge flow rate	Steady-state
U8	Liquid flow to stabilizer	Dynamic only (level control)
U9	Stabilizer reflux flow rate	Steady-state
U10	Stabilizer condenser duty	Dynamic only (level control)
U11	Stabilizer distillate flow rate	Steady-state
U12	Stabilizer reboiler duty	Steady-state
U13	Stabilizer bottoms flow rate	Dynamic only (level control)
U14	Benzene column reflux flow rate	Steady-state
U15	Benzene column distillate flow rate	Dynamic only (level control)
U16	Benzene column reboiler duty	Steady-state
U17	Benzene column bottoms flow rate	Dynamic only (level control)
U18	Toluene column reflux flow rate	Steady-state
U19	Toluene column condenser duty	Steady-state
U20	Toluene column distillate flow rate	Dynamic only (level control)
U21	Toluene column reboiler duty	Steady-state
U22	Toluene column bottoms flow rate	Dynamic only (level control)

Table 6: Selected candidate controlled variables for the HDA process (excluding levels).

Y1	A9/A10 flow rate	Y26	Separator liquid outlet toluene mol fraction
Y2	Fresh toluene flow rate	Y27	Separator liquid outlet xylene mol fraction
Y3	Recycle flow rate of toluene	Y28	Gas recycle ethane mol fraction
Y4	Reactor inlet temperature	Y29	Gas recycle propane mol fraction
Y5	Reactor outlet temperature	Y30	Gas recycle benzene mol fraction
Y6	Separator temperature	Y31	Total flow rate of hydrocarbons to the reaction section
Y7	Bentol feed flow rate	Y32	Hydrogen mol fraction in the reactor outlet
Y8	Fresh gas feed flow rate	Y33	Production rate (flow rate in benzene and stabilizer columns)
Y9	FEHE hot side exit	Y34	Production rate (flow rate in the toluene column)
Y10	Steam flow rate at the separator outlet	Y35	Temperature in an intermediate stage of stabilizer column
Y11	Liquid flow rate at the separator outlet	Y36	Temperature in an intermediate stage of benzene column
Y12	Purge flow rate	Y37	Temperature in an intermediate stage of toluene column
Y13	Separator pressure	Y38	Pressure at the top of stabilizer column
Y14	Furnace heat duty	Y39	Pressure at the top of benzene column
Y15	Cooler heat duty	Y40	Pressure at the top of toluene column
Y16	Toluene conversion at reactor outlet	Y41	Benzene mol fraction in stabilizer column sidestream
Y17	Trimethylbenzene conversion at reactor outlet	Y42	Propane mol fraction in stabilizer column sidestream
Y18	Hydrogen / hydrocarbons ratio in the reactor inlet	Y43	Benzene mol fraction in benzene column sidestream
Y19	Recycle gas flow rate	Y44	Benzene mol fraction in benzene column sidestream
Y20	Mixer ethane mol fraction	Y45	Xylene mol fraction in toluene column bottoms
Y21	Mixer propane mol fraction	Y46	Toluene mol fraction in toluene column bottoms
Y22	Separator overhead vapor ethane mol fraction	Y47	Ethylbenzene mol fraction in toluene column bottoms
Y23	Separator overhead vapor propane mol fraction	Y48	Toluene mol fraction in toluene column overhead
Y24	Separator overhead vapor benzene mol fraction	Y49	Xylene mol fraction in benzene column overhead
Y25	Separator liquid outlet benzene mol fraction		

Primary Controlled Variables

In this case study there are three valuable products of the distillation columns: the composition of benzene in the sidestreams of the stabilizer and benzene columns and the composition of xylene in the bottom of the toluene column. As these are the main products of the unit, their specification in these streams must be considered as active constraints.

The composition of toluene in the recycle stream to the unit is also a pre-defined variable, since an increased content of xylenes in the recycle stream demands higher energy consumption, and may also cause an undesired concentration of ethylbenzene in the process. Thus, this composition is also considered to be an active constraint. Just as considered for xylene, it is not interesting to recirculate back the benzene to the reaction section; therefore, in the bottom of the benzene column, benzene composition must be controlled. The bottom stream of the stabilizer column will be manipulated in order to control the concentration of benzene in the bottom. These six specifications for the distillation columns consume six steady state degrees of freedom, thus leaving $15 - 6 = 9$ remaining degrees of freedom.

The following constraints were considered active for the reaction and separation sections: separator temperature, A_9/A_{10} feed flow rate, fresh toluene flow rate, Bentol flow rate, separator pressure, H_2/HC ratio and reactor inlet temperature. These constraints are not degrees of freedom since their values should be set. Consequently, the remaining number of degrees of freedom is: $15 - 6 - 7 = 2$, which significantly reduces the number of possible sets of controlled variables.

Optimization Analysis for Selection of Remaining Degrees of Freedom

The equation that describes the profit function (J) [M\$/year] to be maximized is described as:

$$J = (p_B F_B + p_X F_X + p_f F_f) - (p_{Tol} F_{Tol}$$
$$+ p_{A9A10} F_{A9A10} + p_{Bentol} F_{Bentol}$$
$$+ p_{H2} F_{H2} + p_{fuel} Q_{fuel} + p_{CW} Q_{CW} \tag{1}$$
$$+ p_{Pow} W_{Pow} + p_{Vap} Q_{Vap})$$

Subject to the following constraints:

Reactor inlet hydrogen/hydrocarbon ratio

$$1 \le \frac{H_2}{HC} \le 3 \tag{2}$$

Feed flow rates (fresh toluene, bentol and stream A_9/A_{10})

$$F_{Tol} = 1984 \text{ kg / h} \tag{3}$$

$$F_{A9A10} = 67595 \text{ kg / h} \tag{4}$$

$$F_{Bentol} = 126443 \text{ kg / h} \tag{5}$$

Reactor temperature

$$T_{reactor} = 697 \text{ K} \tag{6}$$

Benzene purity in the sidestream of stabilizer and benzene columns

$$x_{B,estab} \ge 97.00\% \tag{7}$$

$$x_{B,benzene} \ge 99.99\% \tag{8}$$

Xylene purity in the bottom of the toluene column

$$x_X \ge 68.00\% \tag{9}$$

Toluene purity in the recycle stream

$$x_{Tol} \ge 98.00\% \tag{10}$$

Separator inlet temperature

$$T_{in\_sep} = 500 \text{ K} \tag{11}$$

Separator pressure

$$P_{separador} = 224.3 \text{ Pa} \tag{12}$$

All flow rates and concentrations are non-negative variables.

It is considered that all by-products (purge, distillate vapor of stabilizer and benzene column) are sold as fuel. Additionally:

1. p_B, p_X, p_f, p_{Tol}, p_{Bentol}, p_{A9A10}, p_{H2}, p_{fuel}, p_{CW}, p_{Pow} and p_{Vap} are the prices of feed of benzene, xylene, fuel gas, toluene, bentol, A_9/A_{10} fraction, hydrogen, fuel for the furnace, cooling water, power to the compressor, and steam, respectively (see data in Table 7);

2. F_B, F_X, F_f, F_{Tol}, F_{Bentol}, F_{A9A10} and F_{H2} are the flow rates of benzene, xylene, fuel gas, toluene, bentol (mixture of benzene and toluene), A_9/A_{10} fraction, and hydrogen, respectively, Q_{fuel}, Q_{CW} and Q_{Vap} are heat duties of fuel for the furnace, cooling water, and

steam, respectively, and W_{Pow} is the power to the compressor;

3. $Q_{CW} = Q_{CW, cooler} + Q_{CW, T02} + Q_{CW, T03} + Q_{CW, T04}$;

4. $Q_{Vap} = Q_{Vap, T02} + Q_{Vap, T03} + Q_{Vap, T04}$;

5. $F_f = F_{purge} + D_{T02} + D_{T03};$, where F_{purge} is the purge flow rate, D_{T02} is the distillate flow rate of T_{02} and D_{T03} is the distillate flow rate of T_{03};

6. Annual operation of 8150 hours.

Table 7: Economic data for the process, based on Araújo *et al*. (2007b) and ICIS.

p_B	0.256	\$/kg
p_X	0.312	\$/kg
p_{Tol}	0.145	\$/kg
P_{A9A10}	0.072	\$/kg
p_{H2}	1.455	\$/kg
P_f	1.000	\$/kg
p_{fuel}	3.8×10^{-9}	\$/J
p_{CW}	2.22×10^{-10}	\$/J
p_{pow}	5.6×10^{-5}	\$/J
p_{vap}	2.4×10^{-9}	\$/J

The constant setpoint strategy is simple, but will not be optimal, as a result of disturbances. The effect of these factors (or, more specifically the loss) depends on the choice of the controlled variables, and the goal is to find a set of controlled variables in which the loss is acceptable. In order to identify the structure that satisfies that condition, an analysis of the model was carried out using the resources of optimization of Aspen Plus™. The objective function described in Equation (1) was set up with its constraints. Concentration variables with no active con-

straints were pre-selected, because they have a significant impact on the objective function. The optimal value of these variables was calculated and two disturbances were applied to the process (D1 and D2, +10% and -10% A_9/A_{10} feed flow rate, respectively). The decision variables were the concentrations (mol fractions) from Table 6.

Variables defined as optimal from the standpoint of self-optimizing control were those with less deviation from their optimal value, which implies that the constant setpoint policy has the least impact on the profit function. Table 8 presents the results. It should be noted that the present approach is simpler than that performed by Araújo *et al*. (2007b) who calculated the loss with each variable in the assumed sets kept at its nominal optimal setpoint.

From the described methodology, the selected variables should have been: (i) the separator liquid outlet toluene mol fraction and (ii) the mol fraction of xylene in the reactor outlet. However, as the TDAP process includes a disturbance of toluene ahead of the separator, due to a fresh stream in the feed of the benzene column, it was decided to change the first selected variable by another one. Therefore, comparing with the results obtained by Araújo *et al*. (2007b) for the HDA process, who considered the set composed of mol fraction of methane (inert) in the outlet of the mixer and mol fraction of toluene in the outlet of the quencher, the mol fraction of ethane (inert) at the mixer outlet was considered. With these changes the methodology cannot be considered rigorously self-optimizing. The impacts of this choice are evaluated in the following sections.

Table 8: Effect of disturbances on optimal values of the selected variables.

Candidate Controlled Variable	Nominal Value	Absolute Variation of Nominal Value with D1	Absolute Variation of Nominal Value with D2
Mixer outlet ethane mol fraction	1.004×10^{-6}	4.48×10^{-7}	-4.87×10^{-7}
Mixer outlet propane mol fraction	5.134×10^{-8}	2.33×10^{-8}	-2.59×10^{-8}
Separator overhead vapor ethane mol fraction	8.54×10^{-3}	-2.16×10^{-4}	3.30×10^{-4}
Separator overhead vapor propane mol fraction	4.37×10^{-4}	-5.15×10^{-6}	1.10×10^{-5}
Separator overhead vapor benzene mol fraction	1.02×10^{-4}	1.29×10^{-5}	-1.47×10^{-5}
Separator liquid outlet benzene mol fraction	0.0702032	0.0030892	-0.0035004
Separator liquid outlet toluene mol fraction	0.254397	-0.0017485	0.0023585
Separator liquid outlet xylene mol fraction	0.200455	-0.0099576	0.0101899
Gas recycle benzene mol fraction	1.20×10^{-8}	2.33×10^{-8}	-2.59×10^{-8}
Reactor outlet xylene mol fraction	0.2004806	9.96×10^{-7}	1.02×10^{-7}

DYNAMIC ANALYSIS

Introduction

The dynamic model of the process was obtained from the steady-state model, by using the conversion features of Aspen Plus™ to Aspen Dynamics™. This requires providing information related to the dimensions of the equipment, such as sizes of the columns (level of liquid in the bottom or "sump"), and trays (weir, height, diameter and spacing of the plate) and dimensions of vessels (width and height).

The objective is to perform a dynamic analysis involving the design of the regulatory control layer and the design of the configuration of the supervisory control layer and dynamic simulations with rigorous model of the process to validate the proposed control structure.

The process studied in this work presents more realistic features in relation to the HDA process. The presence of several reversible reactions causes non-linearities of the components to variations in reactor temperature. Figure 3 illustrates this behavior. It is possible to note that only the concentration of xylene has no inversion in the gain relative to the reactor feed temperature, but it also presents a significant gain variation in the range of 450 to 500 K similarly to the other components.
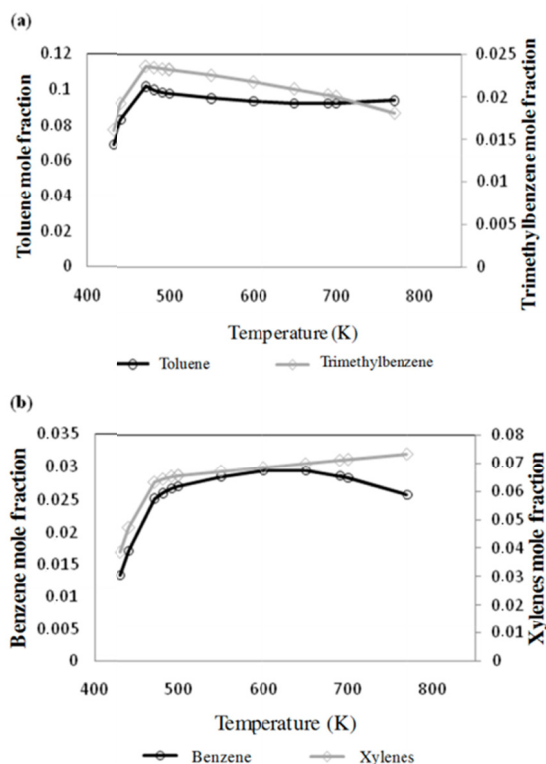

Figure 3: Variation in composition (a) of the reactants and (b) of the products with the reactor inlet temperature.

Strategy 1 − Strategy Oriented to the Control Structure

The procedure proposed by Araújo *et al*. (2007b) was adapted in this present work from the HDA process to the TADP process. The items in the first stage of the procedure were defined during the analysis above and the following items are the second part, the "Bottom-Up" analysis.

Design of the Regulatory Control Structure

The main objective of the regulatory layer is to guarantee a smooth operation, following the set-points provided by the upper layers of the control structure. Usually it is a decentralized control system that maintains a set of output variables at given setpoints. Typically these variables are pressure, levels and some temperatures.

Stabilization of Unstable Modes

In the reaction section one temperature must be controlled in order to stabilize the reactor operation. In the study by Araújo *et al*. (2007b), it was chosen to control the inlet temperature of the reactor by the manipulation of the heat duty of the furnace, which has a direct effect on the temperature.

The liquid levels in the separator and the reflux drum and the sump drum of the distillation columns should also be stabilized. Here it was chosen to manipulate the output flow rates to control these variables. The exception is the level of the reflux drum of the stabilizer column which is controlled by the condenser heat duty due to the absence of distillate liquid.

The dynamic behavior of pressure is usually fast, so the effective control of the pressure in the process is important to stabilize the gas/vapor inventories. First the pressure must be controlled at some point in the reactor recycle loop. As in the TADP process, the separator vessel pressure is an active constraint and this is controlled, rather than the pressure in the reactor as done in Araújo *et al*. (2007b); however the manipulated variable remains the flow rate of purge gas. In each distillation column, pressure is controlled by manipulating the heat duty of the condenser. The exception is the stabilizer column, where the flow of benzene in the sidestream is used as manipulated variable, since the heat duty is used to control the level of the condenser.

Temperature Control

Since the operation of the separator has great impact both in recycle gas separation and in the distil-

late section, its temperature should be controlled. The manipulated variable chosen in this case is the heat duty of the condenser.

The composition control of distillation columns in general is slow due to liquid and gas transport delays as well as large liquid holdup along the column, and the measurement is time consuming and more expensive than temperature control. Therefore, the temperatures are also controlled in the distillation columns in order to avoid fluctuations on a short time scale. The Slope Criterion method (Luyben, 2006) was used. The procedure for selecting the control tray in this method consists of analyzing the temperature profile at the steady state and finding the location in the column that exhibits the largest change in temperature from one stage to another. It was applied and the selected tray for temperature control of T-02 was number 4; number 36 for T-03, and number 46 for control of T-04.

Design of Supervisory Control Structure

The purpose of the supervisory layer is to keep the primary controlled variables in their optimal setpoints. The supervisory layer was adapted from

Araújo *et al*. (2007b). In addition to the previously established composition controllers, an anticipatory control to calculate the ratio H_2/HC is also needed, since this is an active constraint. In the steady-state analysis, the control of the composition of xylene at the outlet of the reactor (by manipulating the setpoint of reactor inlet temperature controller) and the control of ethane in the mixer output by manipulating the flow of recycle gas were defined. Table 9 summarizes all the control loops defined in Strategy 1. The supervisory control structure − composed of the primary xylene composition controller (R-01_CC cascaded with the temperature controller R-01_TC); the ethane composition controller V-01_CC, and the feedforward controller − is shown in Figure 4 that presents the control structure for this strategy.

Tuning of the Controllers

Only P (proportional) control was adopted for liquid levels; for other control loops, PIs (proportional-integral) controllers were employed. The conservative Tyréus-Luyben (Luyben *et al*., 1998, 1997) tuning rules ($K_c = K_u/2.2$; $\tau_I = 2.2\,P_u$; K_u and P_u are respectively the critical gain and period) were used.

Table 9: Summary of the control loops of Strategy 1.

| Tag	Manipulated variable	Controlled variable	K_c (%/%)	τ_I (min)
R-01_TC	F-01 heat duty	R-01 inlet temperature	17.15	3.96
V-01_TC	PREAC cooler heat duty	V-01 temperature	1	20
V-01_PC	V-01 purge flow rate	V-01 pressure	20	12
V-01_LC	V-01 output flow rate	V-01 level	10	-
V-01_CC	Recycle gas flow rate	M-01 output ethane concentration	1	20
R-01_CC	Reactor temperature	R-01 output xylene concentration	1	20
T02_CondPC	T-02 sidestream flow rate	T-02 pressure	44.19	85.8
T02_DrumLC	T-02 condenser heat duty	T-02 reflux drum level	2	-
T02_CC01	T-02 reflux flow rate	T-02 sidestream toluene concentration	0.42	11.88
T02_TC	T-02 reboiler heat duty	T-02 temperature control tray	26.21	19.8
T02_CC02	T-02 temperature control tray	T-02 bottom benzene concentration	0.11	11
T02_SumpLC	T-02 bottom flow rate	T-02 reboiler level	2	-
T03_CondPC	T-03 condenser heat duty	T-03 pressure	98.99	2.64
T03_DrumLC	T-03 sidestream flow rate	T-03 reflux drum level	2	-
T03_CC01	T-03 reflux flow rate	T-03 sidestream benzene concentration	1	20
T03_TC	T-03 reboiler heat duty	T-03 temperature control tray	57.4	6.6
T03_CC02	T-03 temperature control tray	T-03 bottom benzene concentration	0.21	2.59
T03_SumpLC	T-03 bottom flow rate	T-03 reboiler level	20	-
T04_CondPC	T-04 condenser heat duty	T-04 pressure	52.88	5.28
T04_DrumLC	T-04 distillate flow rate	T-04 reflux drum level	20	-
T04_CC01	T-04 temperature control tray	T-04 distillate xylene concentration	2.16	5.28
T04_TC	T-04 reflux flow rate	T-04 temperature control tray	0.73	4.92
T04_CC02	T-04 reboiler heat duty	T-04 bottom toluene concentration	860.6	2.64
T04_SumpLC	T-04 bottom flow rate	T-04 reboiler level	2	-
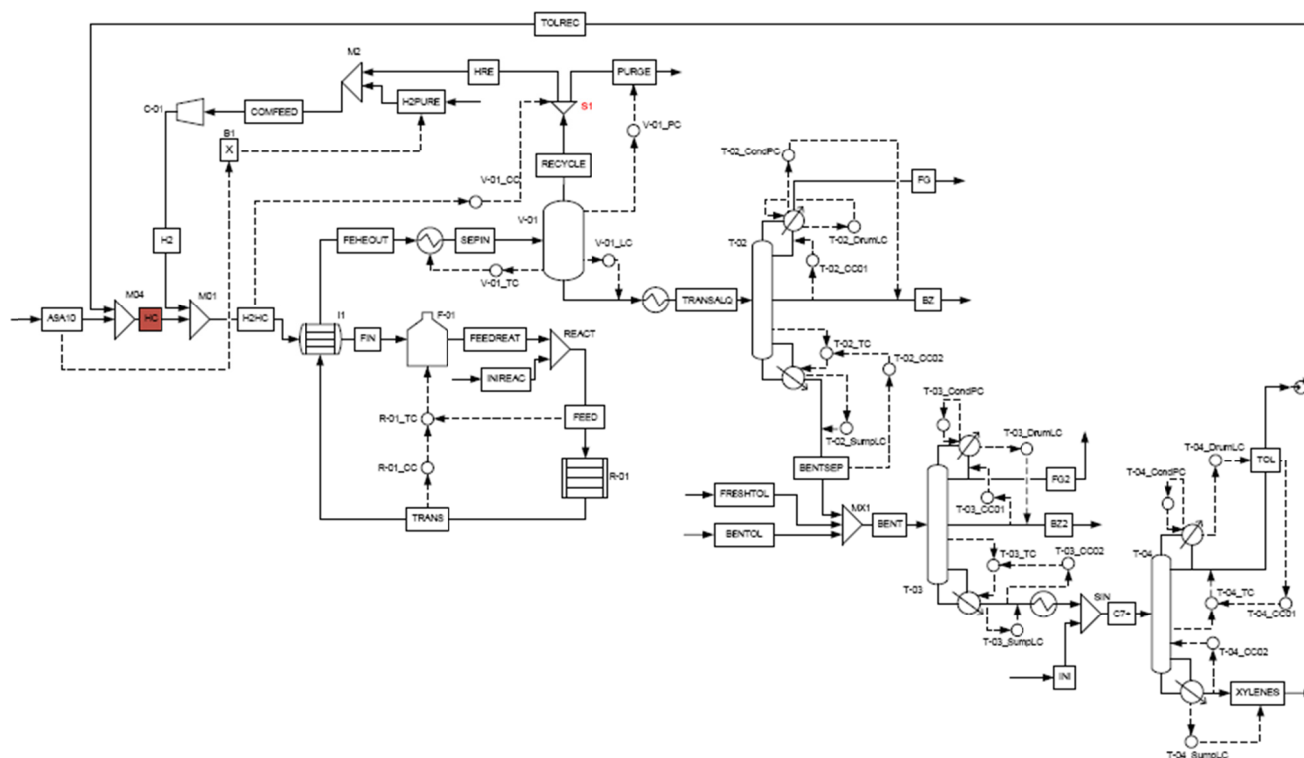
Figure 4: Control structure of Strategy 1.

Strategy 2 – Strategy Oriented to the Process

In the methodology of Luyben *et al*. (1998), the recycles should be fixed in order to avoid the "snowball" effect. In addition, composition controllers are considered necessary only for sale products. In this case the products were considered to be the sidestreams of stabilizers and benzene columns and the bottom of the toluene column. The nine-step procedure is used for obtaining the control structure.

Steps 1 and 2: Control objectives and analysis of degrees of freedom as made previously;

Step 3: The inlet temperature of the reactor should be controlled by manipulating the fuel in the furnace;

Step 4: The feed flow A9/A10 that determine the production rate is fixed;

Step 5: The quality of benzene can be affected mainly by two components: propane and toluene. The separation of propane and benzene in the stabilizer column helps to prevent this by using a temperature controller to set the boilup of the column. The separation of benzene and toluene in the benzene column is easy. As a result, one can establish the boilup of the column by controlling the temperature of the most sensitive tray. To achieve the quality control of the desired product, an on-line analyzer of the top composition is used to adjust the setpoint of the temperature controller. The bottom stream of the toluene column is also considered a saleable stream because it contains xylene product. As this stream carries all the heavy components generated or not reacted in the process, the main purpose of composition control is to prevent that the toluene from the top, which should be recycled to the process, exits the bottom of the column, causing an increased energy consumption in the sections of purification of xylene;

Step 6: The toluene recycle flow rate should be fixed and four pressures must be controlled: in the three distillation columns and in the gas loop. In the stabilizer column, the flow rate of vapor product is the manipulated variable that directly affects the column pressure, but this configuration does not generate good results in terms of separation (Klafke, 2011), so the flow rate of the sidestream is used as manipulated variable. In the benzene and toluene columns, pressure control can be done by manipulating the flow rate of cooling water for adjusting the rate of condensation at the top. In order to maximize the yield of the reactor, the valve in the top of the separator shall be opened and the compressor shall be operated at maximum recycle gas. Therefore, there are two options to control the gas pressure loop: the purge flow or the fresh hydrogen. Once pressure indicates the inventory of hydrogen in the gas recycle loop, the flow of fresh hydrogen is chosen.

Seven liquid levels are presented in the process: the separator level and two levels (reflux and sump

drums) in each column. The most direct way to control the level of the separator is with the flow of fluid into the stabilizer column. The level of the reflux drum of the stabilizer column is controlled by the flow rate of cooling water and the level of the sump drum by the withdrawal of bottom stream. In the benzene column, the flow rate of sidestream controls the level of the reflux drum and the sump is controlled by the bottom flow rate.

Unlike the other columns, the boilup rate of the toluene column is high so that the content of xylene in the bottom of the column is small compared to the recycle of toluene. Therefore, the reboiler steam flow rate was chosen to control the level in the sump drum, which has greater effect. If the flow rate of distillate in the toluene column was used to control the reflux drum level, then the flow rate of liquid in the toluene recycle stream would vary, which is not desired. Therefore the flow rate of fresh toluene is chosen to control this level since it represents the inventory of toluene in the process. This scheme limits large flow variations in the refining section and ensures the component balance for toluene;

Step 7: Ethane is purged from the recycle gas loop to prevent its accumulation and its composition can be controlled by the flow of purge. The heavy components (A_8^+) generated or not reacted are removed by the bottom stream of the toluene column,

where the steam flow controls the level of the sump drum. The benzene inventory is accounted for via the temperature and level controls of the reflux drum in the benzene column and via the temperature and pressure controls in the stabilizer column. The inventory of toluene is accounted for through the level control of the reflux drum in the toluene column. The purge flow and pressure control of the gas loop take over the inventory of hydrogen;

Step 8: At this point the control loops for the individual units can be established, the flow rate of cooling water is defined as manipulated variable in the separator temperature control.

Step 9: The basic regulatory strategy is established. In this step there is freedom to select setpoints of the controllers to optimize cost and performance of the plant. The flow rates of reflux into the columns should be determined based on the energy requirements of the columns and the potential loss of products as contaminants. The selection of setpoints comes from the steady-state analysis.

The final structure can be viewed in Figure 5. Table 10 presents the implemented control loops. Comparing Tables 9 and 10, it can be noted that Strategy 1 presents four more control loops whose controlled variables are concentrations (benzene in the bottom streams of the stabilizer and benzene column; xylene in the distillate of the toluene column and in the
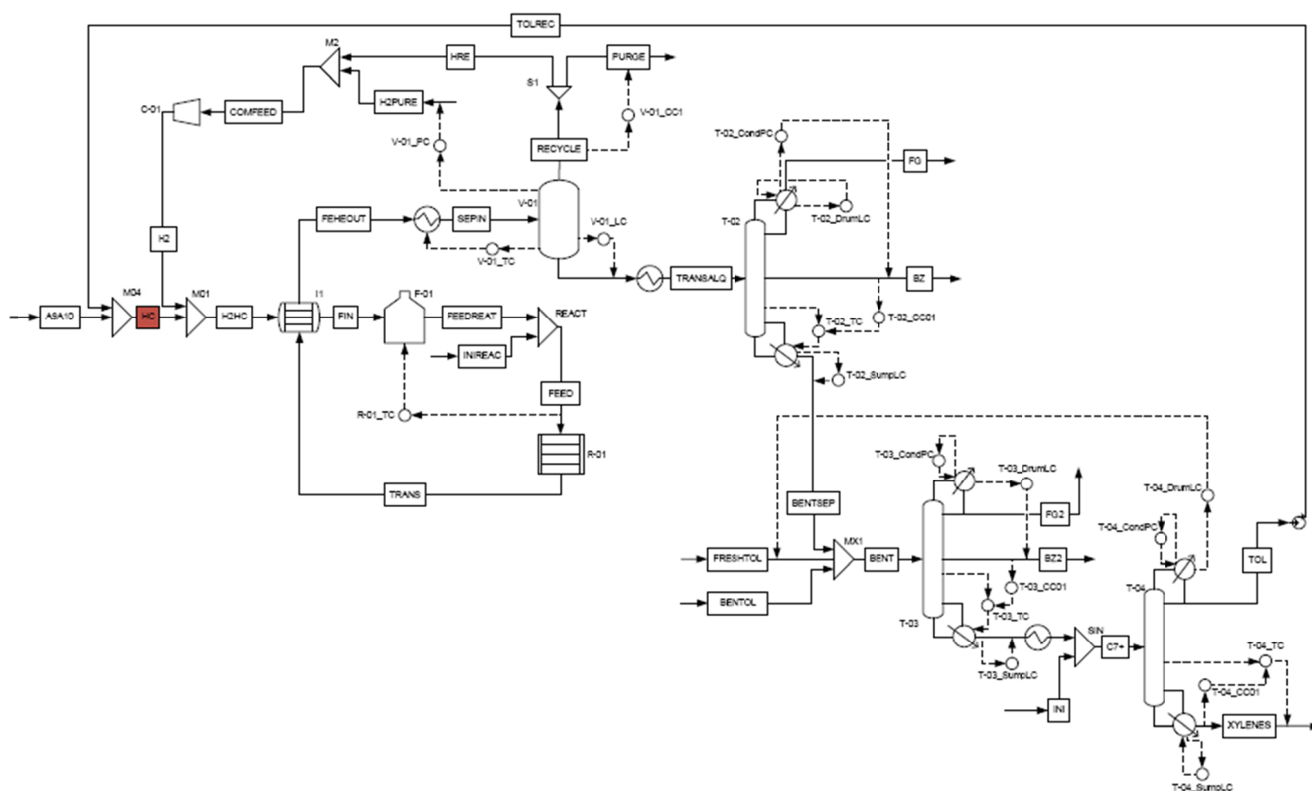

Figure 5: Control structure of Strategy 2.

Table 10: Summary of the control loops of strategy 2.

Tag	Manipulated variable	Controlled variable	K_c (%/%)	τ_I (min)
R-01_TC	F-01 heat duty	R-01 inlet temperature	17.15	3.96
V-01_TC	PREAC cooler heat duty	V-01 temperature	1	20
V-01_PC	Pure Hydrogen Flow	V-01 pressure	20	12
V-01_LC	V-01 output flow rate	V-01 level	10	-
V-01_CC	V-01 purge flow rate	V-01 ethane concentration in the top output	1	20
T02_CondPC	T-02 sidestream flow rate	T-02 pressure	44.19	85.8
T02_DrumLC	T-02 condenser heat duty	T-02 reflux drum level	2	-
T02_CC01	T-02 temperature control tray	T-02 sidestream toluene concentration	0.42	11.88
T02_TC	T-02 reboiler heat duty	T-02 temperature control tray	44.2	85.8
T02_SumpLC	T-02 bottom flow rate	T-02 reboiler level	2	-
T03_CondPC	T-03 condenser heat duty	T-03 pressure	98.99	2.64
T03_DrumLC	T-03 sidestream flow rate	T-03 reflux drum level	2	-
T03_CC01	T-03 temperature control tray	T-03 sidestream benzene concentration	1	20
T03_TC	T-03 reboiler heat duty	T-03 temperature control tray	57.4	6.6
T03_SumpLC	T-03 bottom flow rate	T-03 reboiler level	20	-
T04_CondPC	T-04 condenser heat duty	T-04 pressure	52.88	5.28
T04_DrumLC	Fresh toluene flow rate	T-04 reflux drum level	20	-
T04_CC01	T-04 temperature control tray	T-04 bottom toluene concentration	0.32	22.44
T04_TC	T-04 bottom flow rate	T-04 temperature control tray	6.12	17.16
T04_SumpLC	T-04 reboiler heat duty	T-04 reboiler level	2	-

output of the reactor). The effects of these additional loops will be appreciated through dynamic simulation in the next item.

Evaluation of the Proposed Control Structures

The two control structures were evaluated using Aspen Dynamics™ and considering a +10% disturbance in the feed flow rate of the A_9/A_{10} stream. This disturbance was selected because increasing the heavy aromatic content of the TADP feed is an inter-

esting condition from the economic point of view of the aromatic complex, since this is a stream of low commercial value with the potential to turn into products of high value such as benzene and xylenes. The results are presented in Figure 6 to Figure 9.

Figure 6 presents the behavior of the regulatory temperature control loops of the reaction section. The setpoint of the reactor temperature in Strategy 1 decreases due to the actuation of the xylene concentration controller (R-01_CC). Figure 7 exhibits the results for the structure of the supervisory layer

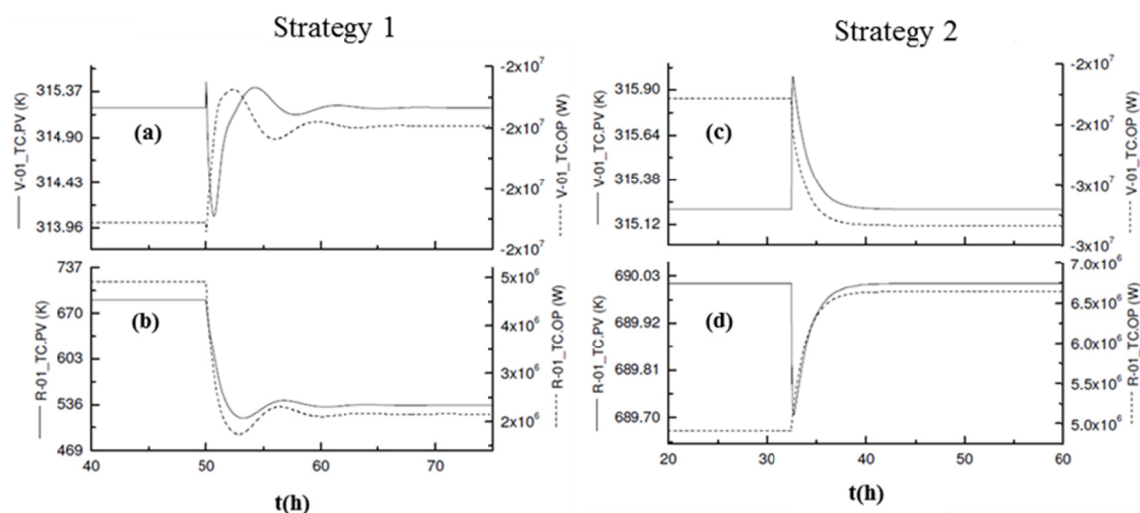

Figure 6: Reaction section regulatory control under disturbance of 10% in A_9/A_{10} flow rate. (a) and (c) – variation in separator temperature (V-01_TC. PV) with the manipulation of cooler heat duty (V-01_TC. OP). (b) and (d) – variation in reactor temperature (R-01_TC. PV) with the manipulation of furnace heat duty (R-01_TC. OP). Controlled/manipulated variables (solid/dashed lines).
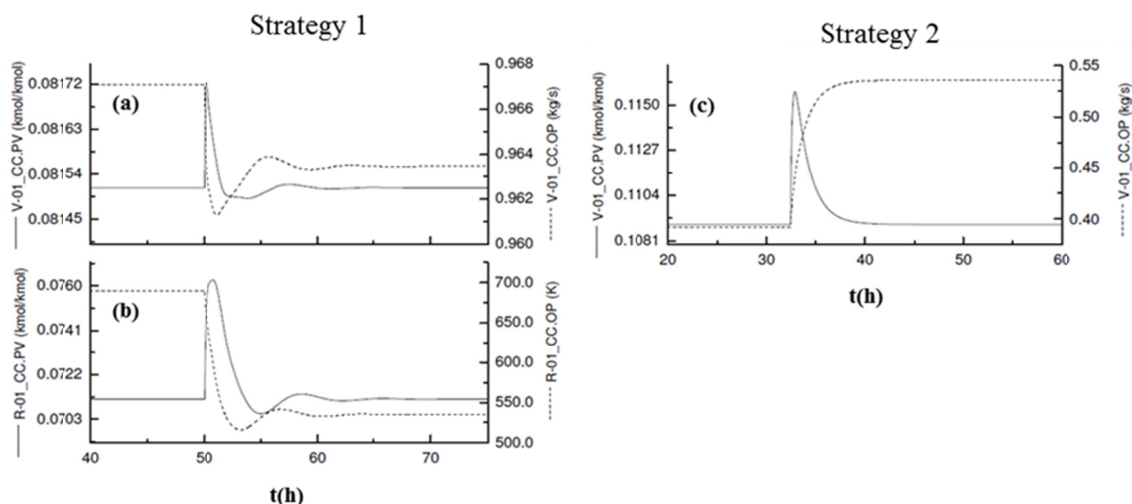
Figure 7: Reaction section supervisory control under disturbance of 10% in A_9/A_{10} flow rate. (a) – variation of ethane concentration in the mixer outlet (V-01_CC. PV) with the manipulation of recycle gas flow rate (V-01_CC.OP) for strategy 1. (b) – variation of xylene concentration in reactor outlet (R-01_CC.PV) with the manipulation of the setpoint of reactor temperature (R-01_CC.OP) for strategy 1; (c) – variation of ethane concentration in the overhead of separator (V-01_CC.PV) with the manipulation of purge flow rate (V-01_CC. OP) for strategy 2. Controlled/manipulated variables (solid/dashed lines).

proposed for the reaction section. The control of ethane concentration is performed by manipulating the flow rate of recycle gas in Strategy 1 (Figure 7a) and the purge flow rate in Strategy 2 (Figure 7c). Strategy 1 also presents one more controller, the reactor output xylene concentration controller (Figure 7b), that justifies the difference in the behavior of the reactor temperature (Figure 6b and d).

The stabilizer column absorbs the impacts of the disturbances and these are minimized by the controllers of benzene and toluene columns for both strategies. Strategy 1 presents one more controller in this column, which is the bottoms composition controller. Figure 8 shows that in both strategies the operational constraint of benzene composition in the sidestream, Equation (7), was not satisfied, although the second strategy presented a lower loss of purity. The control of the benzene composition with the reflux flow rate in Strategy 1 presents a more non-linear behavior, as it sometimes must act in order to prevent contamination with toluene and other times with propane. Additionally, the variables chosen with constant setpoint did not guarantee optimal behavior for the disturbance in the A9/A10 feed stream flow rate.

Results for the benzene and toluene columns are not shown here due to space reasons. For both columns and both strategies, the impacts of the disturbance are effectively absorbed (Klafke, 2011).

Strategy 3 – Modification in the Strategy 1

Strategy 1 presented difficulties to comply with the constraint of the benzene composition in the sidestream of T-02. Although this is not a stream of benzene product intended for sale outside the aromatic complex, increasing the content of impurities, especially light gases, corresponds to a significant disturbance in the adjacent consumer unit, so the minimum purity is an important constraint.

The composition control of the sidestream of the benzene column, more specifically of the propane mol fraction, is accomplished by manipulating the flow rate of distillate vapor. The reflux flow rate in this configuration is kept constant. It is important to note that this solution is only plausible due to the fact that this column is a high reflux ratio type, which ensures that variations in the flow of distillate vapor do not significantly alter the flows inside the column. Changes in control structure of T-02 in relation to Strategy 1 are shown in Figure 9 and Table 11, resulting in the Strategy 3.
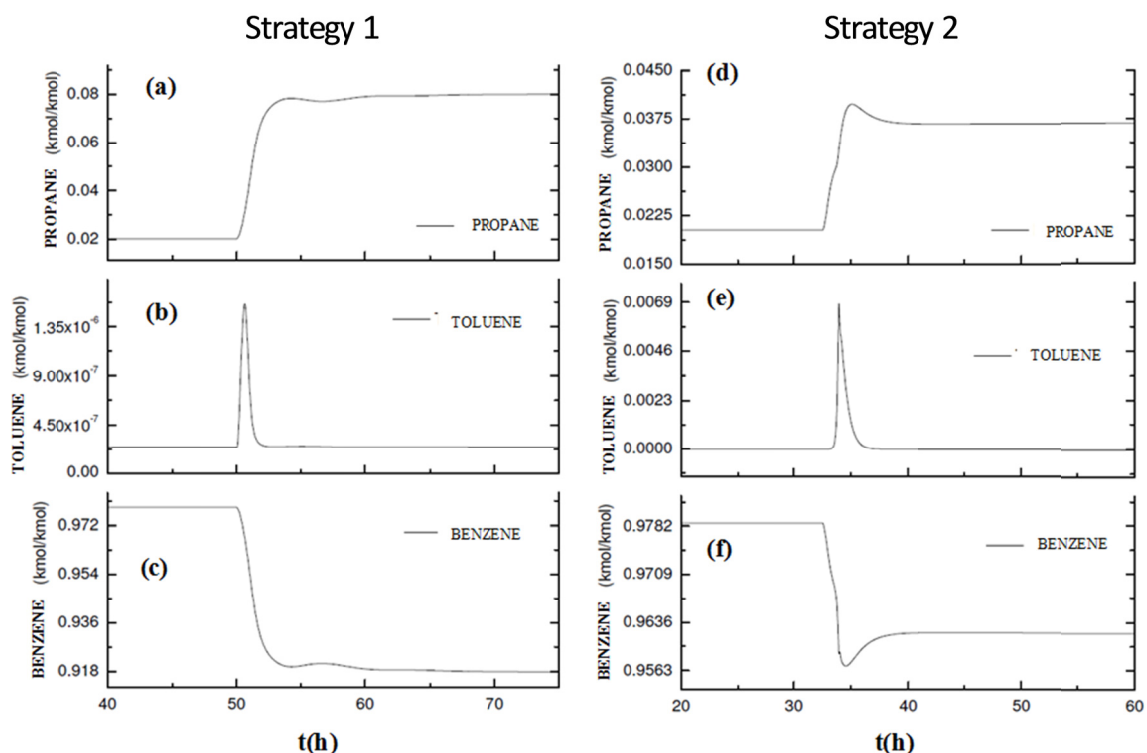
Figure 8: Sidestream composition in T-02 with disturbance of 10% in A_9/A_{10} feed stream flow rate.
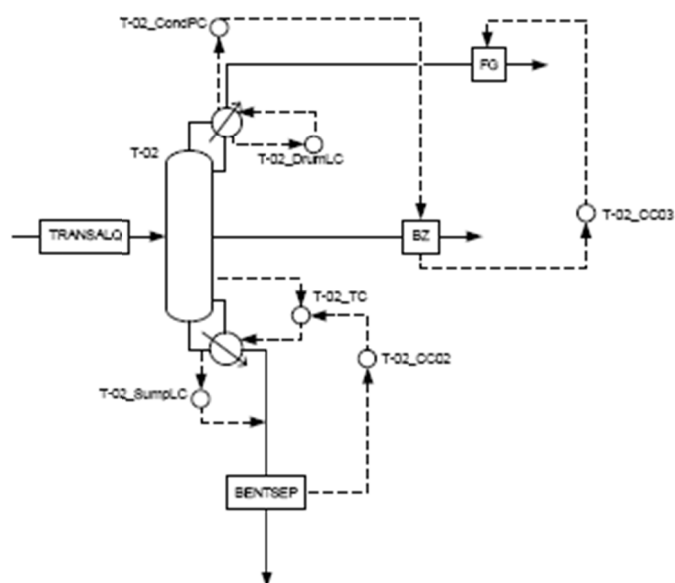
Figure 9: Proposed modification in Strategy 1 control structure of T-02.

Table 11: Summary of the control loops of Strategy 3.

Tag	Manipulated variable	Controlled variable	K_c (%/%)	τ_I (min)
T02_CondPC	T-02 sidestream flow rate	T-02 pressure	44.19	85.8
T02_DrumLC	T-02 condenser heat duty	T-02 reflux drum level	2	-
T02_CC03	T-02 fuel gas output flow rate	T-02 side stream propane concentration	0.42	11.88
T02_TC	T-02 reboiler heat duty	T-02 temperature control tray	26.21	19.8
T02_CC02	T-02 temperature control tray	T-02 bottom benzene concentration	0.11	11
T02_SumpLC	T-02 bottom flow rate	T-02 reboiler level	2	-

The results presented in Figure 10 indicate that the proposed regulatory control is effective and the constraint in the benzene composition was complied. The change in Strategy 1 which generated Strategy 3 was also effective through the economic aspects since the profit J obtained with Strategy 3 was 2.8 MM$/year against 2.3 MM$/year for Strategy 2.

Strategy 3

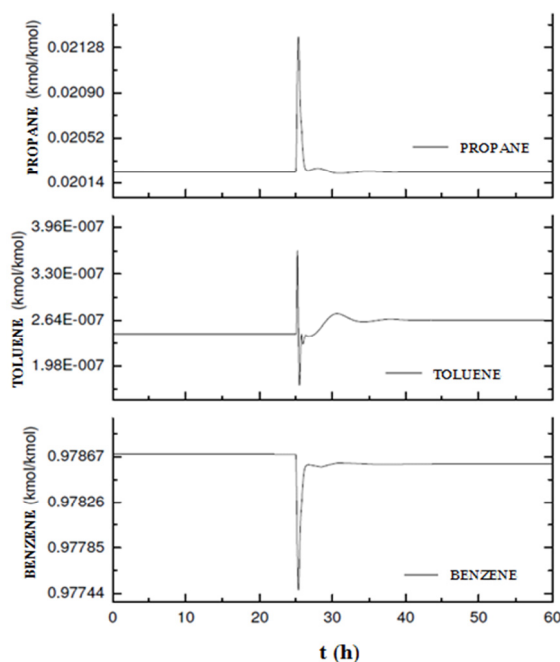**Figure 10:** Sidestream composition in T-02 with disturbance of 10% in A_9/A_{10} flow rate for strategy 3.

CONCLUSIONS

The HDA process has become obsolete with the development of a new technology capable of processing heavy aromatic components, adding value to raw materials, and producing fewer unwanted by-products. Although this process has associated commercial names, here it is called TADP, as in Das *et al.* (1993). This work was based on two methods of plantwide control - the control structure approach (based on Araújo *et al.* (2007b)) and the process-oriented approach (based on Luyben *et al.* (1998)) – originally defined for the HDA process and used here to develop control structures for the TADP process. This process unit was inserted into an aromatic complex and effects of the interaction between the units have been discussed.

The initial results obtained in this work showed that, in terms of control performance, both strategies are similar. However, when a disturbance of 10% in the heavy aromatic feed was applied, the modified

Strategy 1 did not guarantee optimal behavior for this disturbance and for the set of variables chosen for constant nominal set-point. Because of that, changes in the configuration of the stabilizer column control were carried out and better result in terms of composition control and economic performance were obtained. So, it is considered that future plantwide control implementations aiming at the improvement of the profit of the TADP unit must be sought.

The TADP process, approached from the plant-wide point of view for the first time in this work, can be used as a new challenge for problem-control studies, replacing the conventional HDA process, being technologically more up-to-date and more challenging. Its complexity and economical importance make it a promising scenario where more recent process control strategies may be tested, such as Economic Model Predictive Control (EMPC) (Ellis *et al.*, 2014) or Distributed Model Predictive Control (Christophides *et al.*, 2013).

NOMENCLATURE

An	Aromatic hydrocarbon compound with n carbons
An^+	Cut composed of aromatic hydrocarbons with n carbon atoms of the lighter component
An^-	Cut composed of aromatic hydrocarbons with n carbon atoms of the heavier component
Cn	Hydrocarbon compound with n carbon atoms
Cn^+	Cut composed of hydrocarbons with n carbon atoms of the lighter component
Cn^-	Cut composed of hydrocarbons with n carbon atoms of the heavier component
c	Controlled variable
u	Manipulated variable
d	Disturbance variable
c_s	Controlled variable reference value
J	Objective function
K_c	Proportional gain
τ_I	Integral time constant

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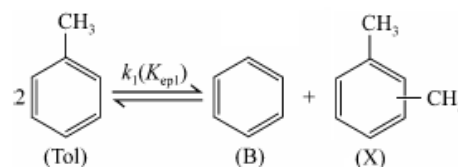
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APPENDIX

Kinetics of Reactions of the TADP Process

The kinetic model (Ouguan *et al*., 2007) is presented below. In this model, *r* is the reaction rate, *a* is the mass fraction (%), ϕ is the function of catalyst deactivation which is assumed as uniform for all reactions and equal to 1 in this work, *k* is the kinetic constant of reaction and *K* is the chemical equilibrium constant. The components involved are: toluene (Tol), benzene (B), xylenes (X), methylbenzene (MB), ethylbenzene (EB), propilbenzene (PB), trimethylbenzene (TMB), and $C_{10}A$.

Disproportionation of Toluene (Reversible)

$$r_1 = k_1 \cdot \phi \left(a_{Tol}^2 - \frac{a_B \cdot a_x}{K_1} \right) \qquad (A1)$$

$$r_2 = k_2.\phi\left(a_{Tol}^2 - \frac{a_B.a_{EB}}{K_2}\right) \qquad (A2)$$

$$r_7 = k_7.\phi\left(a_{TMB}^2 - \frac{a_X.a_{C_{10}A}}{K_7}\right) \qquad (A7)$$

Transalkylation of Toluene with Trimethylbenzene (Reversible)

(Tol)   (TMB)   (X)

$$r_3 = k_3.\phi\left(a_{Tol}.a_{TMB} - \frac{a_X.a_X}{K_3}\right) \qquad (A3)$$

(Tol)   (TMB)   (EB)

$$r_4 = k_4.\phi\left(a_{Tol}.a_{TMB} - \frac{a_{EB}.a_{EB}}{K_4}\right) \qquad (A4)$$

Dealkylation (Irreversible)

(MEB)   (Tol)

$$r_5 = k_5.\phi.a_{MEB} \qquad (A5)$$

(PB)   (B)

$$r_6 = k_6.\phi.a_{PB} \qquad (A6)$$

Disproportionation of Trimethylbenzene (Reversible)

(TMB)   (X)

The kinetic constant of the ith reaction is defined by the Arrhenius equation:

$$k_i = k_{0i}.\exp\left(-\frac{E_{ai}}{RT}\right), \qquad (i = 1-7) \qquad (A8)$$

The chemical equilibrium constants are calculated as follows, for reactions 1 to 4 and 7, where M_c is the molecular weight of the component C. The thermodynamic constant K_{epi} is given by Equation (A14).

$$K_1 = K_{ep1}.\frac{M_B.M_X}{M_{Tol}^2} \qquad (A9)$$

$$K_2 = K_{ep2}.\frac{M_B.M_{EB}}{M_{Tol}^2} \qquad (A10)$$

$$K_3 = K_{ep3}.\frac{M_X^2}{M_{Tol}.M_{TMB}} \qquad (A11)$$

$$K_4 = K_{ep4}.\frac{M_{EB}^2}{M_{Tol}.M_{TMB}} \qquad (A12)$$

$$K_7 = K_{ep7}.\frac{M_X^2.M_{C_{10}A}^2}{M_{TMB}^2} \qquad (A13)$$

$$K_{epi} = \exp\left(-\frac{\Delta G_i}{RT}\right), \qquad (i = 1-4, 7) \qquad (A14)$$

The kinetic parameters are presented in Table A1.

Table A1: Kinetics parameters of the model, based on Ouguan *et al*. (2007).

Reaction	Direct reaction		Reverse reaction	
	k_{0i}, s^{-1}	E_{ai}, kJ mol^{-1}	k_{0i}/K, s^{-1}	$(E_{ai})_{inv} = E_{ai} - \Delta G$, kJ mol^{-1}
1	9.106 x 10^{10}	102	3.269 x 10^{11}	94.8
2	9.534 x 10^{13}	150	1.567 x 10^{15}	134.1
3	5.712 x 10^{10}	96.77	1.273 x 10^{11}	92.07
4	6.001 x 10^{13}	150	2.804 x 10^{15}	127.8
5	6.126 x 10^{10}	101.8	-	-
6	3.263 x 10^{10}	95.8	-	-
7	3.213 x 10^{9}	106	1.338 x 10^{11}	84.7