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AN APPROACH TO OPTIMIZATION OF A THREE PHASE CATALYTIC SLURRY REACTOR BY EVOLUTIONARY OPTIMIZATION WITH GENETIC ALGORITHMS

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Abstract: The present work introduces an investigation study about the optimization of a three-phase catalytic slurry reactor with phase changes. The study aims to determine the optimal operational conditions and, in a second stage, advanced control algorithms will be evaluated. A dynamic heterogeneous mathematical model formulation was used and genetic algorithms (GA) were employed to solve a nonlinear quadratic problem subject to bounds on the variables. The solutions obtained using the genetic algorithms gave suitable results to optimize the process by adjusting parameters as the reactor feed temperature and the flow rate of one of the reactants. It was possible to reach operation conditions that ensured a suitable profit and an acceptable compromise with the specifications of the desired product.

Keywords: optimization, two-layer approach, genetic algorithms, advanced control, dynamic modeling and hydrogenation.

1. INTRODUCTION

On-line optimization must cope with the variability of the process conditions, originated by disturbances that significantly affect the process economy. On the one hand, there are internal disturbances caused by intrinsic changes in the unit conditions, mainly process degradation. On the other hand, there are external disturbances due to changes in material and energy streams that enter the process (besides the information flows, e. g. the economical parameters).

Decision variables (or manipulated) used for the optimization must be selected considering the operation conditions, which are subjected to a narrow range of values called the "operability range" due to limitations and stability of the process. Moreover, it is a challenge to conciliate optimization with an appropriate computation effort when considering real time process integration.

Several factors can influence the dynamic behaviour of multiphase catalytic reactors such as reactant and coolant temperatures, concentrations, environmental restrictions, phase change and variations of physico-chemical properties. The present work introduces an investigation study about the optimization of a three-phase catalytic slurry reactor with phase changes. The study aims to determine the optimal operational conditions and, in a second stage, advanced control algorithms will be evaluated. A dynamic heterogeneous mathematical model formulation was used, which basically consists on mass and energy balance equations for the reactants as well as for the catalyst particles involving phase change of both reactants and coolant.

Thus, to accomplish the objective of this work, Genetic algorithms (GA) were used to solve the optimization problems. It is presented a real time process integration involving the optimization and control of the process illustrated in Figure 1 (Melo et al., 2005), with the two-layer approach. This is a hierarchical control structure where an optimization layer calculates the set-points to the advanced controller, which is based on the Dynamic Matrix Control (DMC) procedure.

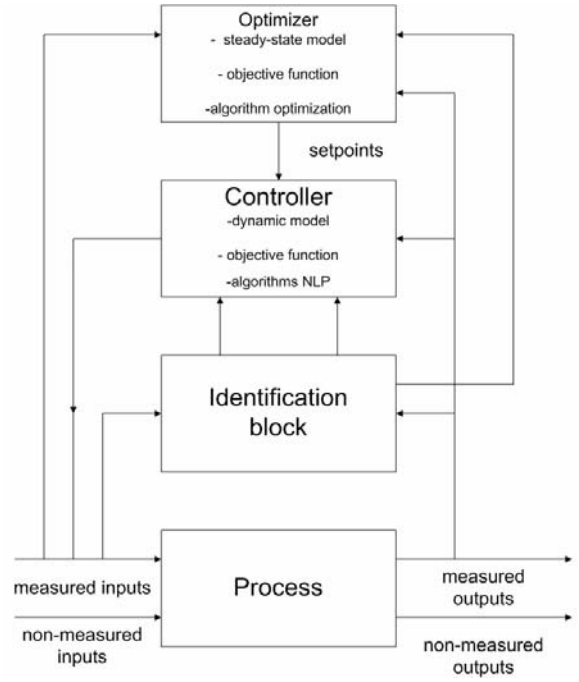

Fig. 1. Two-layer approach.

2. DYNAMIC MODEL USED TO REPRESENT THE MULTIPHASE CATALYTIC REACTOR

A dynamic model was formulated aiming to incorporate the main phenomena that take place in three-phase reactors. The model consists of mass and energy balance equations, and also an equation for the refrigerant fluid (coolant). This non-isothermal heterogeneous model includes: the resistances to mass and heat transfers at the gas-liquid and liquid-solid interfaces, as well as for the catalyst particle; the heat exchange with the coolant fluid; and the consideration of the variation of physico-chemical properties as well as their impact on the mass and heat-transfer coefficients that are calculated through correlations available in the literature. Also, the model includes a multicomponent flash to evaluate the effect of phase change of the reacting medium, and an appropriate procedure to correct the heat-transfer global coefficient to incorporate the effect of phase change of the coolant fluid (Mariano et al., 2005).

The partial differential equations (PDEs) were converted to ordinary differential equations (ODEs) by discretization using the global orthogonal collocation method. After the discretization of the spatial coordinates for the catalyst particle and for the reactor, a system of ODEs and LEs (linear equations) are obtained. The numeric integration is used to obtain profiles in time.

The model was formulated focusing on hydrogenation reactions that are important reactions of industrial processes. This work considers the hydrogenation of o-cresol on Ni/SiO_2 catalyst (Hichri et al., 1991). However, as there are many other hydrogenation reactions of industrial interest, the software can easily incorporate other reactions due to the generality adopted during the development of the model. The hypotheses adopted during the development of the dynamic model of the tubular reactor are (Mariano et al., 2005):

1. A pseudo-homogeneous media is assumed for the liquid and the solid with respect to solid movement. This means that there is no velocity difference between solid and liquid.

2. Negligible pressure variations.

3. Reaction of the type A (g) + νB (l) → νC(l), taking place in the catalyst and having a kinetic that is dependent on the concentrations of A and B.

4. Phase change happens in the system.

The radial dispersion has been neglected for the fluid phase, which is a very common assumption in multiphase reactors as normally found in literature (Gianetto and Silveston, 1986). The superficial velocities and the hold-ups are updated taking into account the changes in temperature, pressure and concentration. Some discussions on the formulation of dynamic models are presented elsewhere: Julcour et al. (1999), Salmi et al. (2000) and Vasco de Toledo et. al. (2001).

The hydrogenation of o-cresol on Ni/SiO_2 producing 2-methylcyclohexanol is represented by:

$$3H_2(g) + C_6H_4OHCH_3(l) \rightarrow C_6H_{10}OHCH_3(l)$$

or $A\,(g) + \nu B\,(l) \rightarrow \nu C\,(l)$

which is a generic representation of three-phase hydrogenation reactions. In this case, *A* stands for hydrogen, *B* for o-cresol, *C* for 2-methylcyclohexanol and ν is the stoichiometric coefficient (equal to 1/3). The following expression has been obtained for the reaction rate (Hichri et al., 1991):

$$R_A = k\frac{K_A K_B C_A C_B}{(1 + K_A C_A)(1 + K_B C_B)}$$

The constants are functions of temperature according to the Arrhenius law type, and *C*i is the concentration of the component i (kmol/m^3).

In exothermic hydrogenation reactors, an oscillatory behavior may occur during phase change, as follows. To assure the reaction occurrence, both reactants must achieve the catalyst particles. Due to the heat release by the reaction, particle temperature increases, and, depending on the temperature, pressure and the cooling system conditions, the liquid at the catalyst surface may evaporate. When such situation happens, the rate reaction decreases drastically, since the liquid reactant is no longer present at the catalyst surface. As a consequence of

the insignificant reaction advance and of reactor heat removal by the cooling system, reactor temperature decreases, which leads to condensation of the reactant that was previously evaporated, and the necessary conditions for the reaction to occur is once more established. Thus, this phenomenon will cause an oscillatory behaviour on the reactor temperature along its length.

The numerical strategy used to couple the flash calculation to the dynamic model is as follow: during pauses in the integration (fixed time intervals), the bubble and the dew points of the reacting medium or, as another option, the Hanika criteria (Hanika et al., 1976), are evaluated for each axial position of the reactor discretization grid. If the temperature of the reacting medium at each axial position is found between the bubble and dew points or if the criteria are satisfied, the multicomponent flash is calculated according to the classical methodology (Henley and Seader, 1981; Raman, 1985). Consequently, the heat involved in the phase change (Q_{FLASH}) is computed by the energy balance of the fluid phase and the components concentrations are updated, except for the hydrogen, which is non-condensable and is considered to be dissolved in the liquid phase.

The multicomponent flash calculation introduces the F factor, which is proposed to represent the physical phenomenon of phase change in multiphase catalytic reactor. The results obtained with the flash procedure (liquid and vapor phases concentration and Q_{FLASH}) are multiplied by F ($0 \leq F \leq 1$). In this way, F is a correction factor that evaluates the amount of material that really experiments phase change, because the conventional flash calculations alone do not represent the real situation, since the reacting system is too complex. It is known that not all mixture in consideration is flashed: when the total flash is considered (F = 1), the temperature changes observed by the model results are too large to allow convergence or even to be real. In order to validate the model, industrial data and information extracted from an industrial scale hydrogenation reactor, located in a large scale production plant located nearby Campinas (Brazil), were used, and the F-factor value (0.005) for the flash calculation was adjusted according to these data: this value reproduced satisfactorily the variations in the temperature observed in practice (~10ºC).

The dynamic model also takes into consideration the phase change of the coolant fluid, via a correction procedure of the global heat-transfer coefficient (U). The great amount of energy released in hydrogenation reaction may increase the temperature of the coolant fluid until evaporation. For this reason, the flow regime of the coolant fluid can present two situations: monophase (liquid or steam) and two-phases (liquid + steam), having a large number of correlations that predict the convective coefficient. Thus, variations in the heat-transfer coefficient due to evaporation will change the global heat-transfer coefficient, *U*, which may affect the dynamic behavior of the reactor. More details on the correlations and on the equations can be found in Mariano et al. (2005).

3. DYNAMIC PERFORMANCE OF THE MULTIPHASE CATALYTIC REACTOR

This section shows the reactor dynamic behaviour (model presented in session 2) after step perturbations in manipulated variables with the greater influences on the reactor exit temperature, to know, the reactant feed temperature (Tfo) and the coolant temperature (Tr). These variables were select employing an experimental design.

Figures 2 e 3 show the effect of the phase change of the reacting medium in the dynamic behaviour of the reactor after a step perturbation in Tr. The observed responses are, respectively, the reactor exit temperature and the concentration of o-cresol in the liquid phase.

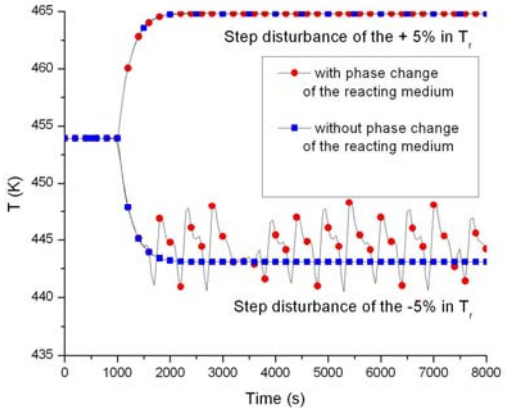

Fig. 2. Effect of the phase change of the reacting medium on the reactor exit temperature.

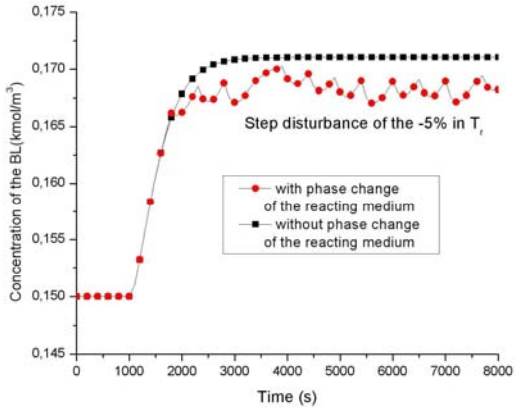

Fig. 3. Effect of the phase change of the reacting medium on the liquid phase o-cresol concentration at the exit of the reactor

The phase change of the reactant fluid gives rise to steep temperature gradients within the reactor due to

the heat involved in the phase change according to the flash calculation. Thus, temperature increases, when condensation occurs, and it decreases as the reacting medium vaporizes. This oscillatory dynamic behaviour is also observed for reagent (o-cresol) and product (2-methyl-ciclohexanol) concentrations.

4. PROPOSED OPTIMIZATION PROBLEM

The optimization study aimed to maximize profits. Due to the complexity of the reactor behaviour, the Genetic Algorithm (GA) is used in this work. It is a method that utilizes the stochastic search nature of genetic algorithms to find combinatorial design within the search space for the solutions.

4.1. Selection of the decision variables

The decision variables used for the optimization of the reactor must be selected among the operation ones. After considering the effect of each of these variables on the objective functions, industry requirements and the easiness of how these variables can be changed in the plant, the following variables were chosen as the decision variables: the feed flow rate of reactant B (FB) and the reactor feed temperature (Tfo). The feed flow rate of reactant A (FA) was kept at $1.5x10^{-2}$ kmol/m^3.

These variables can be either parametric design variables or configuration variables. It is advisable to choose those variables that may have a slight chance of being made common. The primary purpose of this step is to identify only those variables that have a significant impact on the reactor performance in terms of the product, such as flow rate of reactant B and feed temperature.

4.2. Selection of objective functions

The optimization of any industrial process aims, obviously, the profit maximization. Thus, the profit function is a natural choice as an objective function. The profit function (P), as outlined by Cutler and Perry (1983), can be calculated as follows:

P = selling price of the products - cost of raw materials - cost of operating - energy cost.

Then, in this work, the optimization problem was based on the expression designed by Cutler and Perry (1983) and adapted for the multiphase reactor as follows:

$$J = \max P$$

$$P= a*(FA+FB)*XC - b*FA – c*FB - \text{energy cost}$$

where: XC is the molar fractions of the 2-methyl-cyclohexanol; FA is flow rate of reactant A (hydrogen); FB is flow rate of reactant B (o-cresol).

Costs of the raw materials are, in US$/Kg, 0.50 and 0.80, respectively, for o-cresol and hydrogen; the selling price of 2-methylcyclohexanol is US$ 7.50/Kg and, generally, the reagents and products are transported in barrels of 200 Kg or in tank trucks. Thus, the coefficients a, b and c in the function P are 1500, 100 and 160, respectively.

The energy cost related to the steam can be attained by adjusting heating and cooling with negligible cost. Thus, P is given by the following equation:

$$P= 1500.0 * (FA + FB)*XC-100FA-160FB$$

For the operation conditions (FA=$1.5x10^{-2}$ kmol/m^3), and a step disturbance of -5% in Tr (feed coolant temperature), with F=0.005, the optimum operating point corresponds to: FB= $8{,}3368x10^{-3}$ kmol/m^3 and Tfo= 499.9298K, and the profit has a single maximum point = 0.096 US$/s.

The optimal operational point is calculated by an optimizer based on direct search methods, in this case by the genetic algorithms (GA). The optimization involves a process model, ranges and limits of the independent and dependent variables, economic and product quality information, with or without equality and inequality constraints. The process model includes component and overall material balances, thermodynamic and physical property information.

Genetic algorithms are a global optimization based on the principle of “survival of the fittest”. They make an attempt to locate the global optimum solution. However, the ability to successfully identify the global optimum is dependent on various factors. The influence of these factors in the convergence and a few drawbacks of genetic algorithms are described below: the use of a fitness function; no use of any gradient information to capture the global optimum; the search is driven to different areas in the design space based on population modification via crossover or mutation genetic operations; important control parameters of a simple GA include the population size (number of individuals), crossover and mutation rate. The rate of convergence and computation time is dependent on these control parameters and on the type of particular problems.

Several researchers have studied the effects of these parameters on the performance of a GA. Therefore, it is very important to choose suitable control parameters, such as population size, generation size, and crossover and mutation probability. They were chosen based on the work of several researchers and are shown in session 5 (Deb et al., 2002; Carroll, 2004).

The fitness function of the GA dictates how the GA explores the design space to find out better solutions. All possible constraints should be encoded into the

GA so that only feasible designs are chosen for reproductions.

5. RESULTS AND DISCUSSION

The genetic algorithm (GA), designed by Carroll (2004), was run for 10 generations with a population size of 10, i.e., a total of 100 candidate maps were evaluated. This run took around 4 hours using an AMD Athlon processor (2.8 GHz, 768 Mb RAM). Since the computational time demanded to achieve convergence is directly proportional to the necessary number of generations, a maximum number of generations were used as stopping criterion in the genetic programming.

Table 1 gives the bounds of the dependent and independent (decision) variables. The values of the computational parameters used in the GA for solving the optimization problem are 10; 10; 0.5; 0.01; 21 e 2 for maximum number of generations, population size, crossover probability (P_{cross}), mutation probability (P_{mute}), maximum of chromosomes (binary bits) e maximum of parameters (nparmax), respectively.

Table 1. Bounds of the variables used in GA

Parameters	Lower bound	Upper bound
FB (kmol/m^3)	0	$1.49x10^{-3}$
Tfo (K)	450	500

Figure 4 shows the resulting fitness values. They illustrate the convergence of the algorithm over time and show the fitness of both best and average members of the population in each generation. With the increase of the number of generations (10, 15 and 20), better solutions with constant values are obtained, which represent a profit increase.

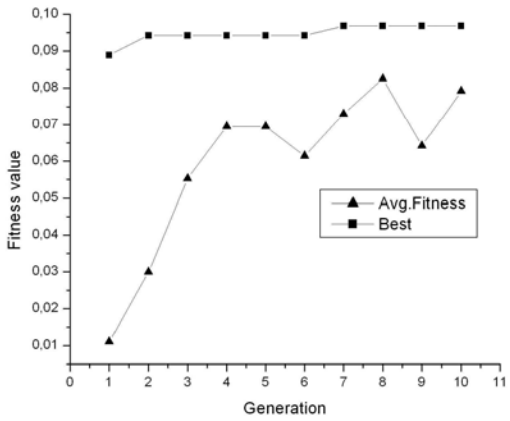

Fig. 4. Fitness values over time for the genetic algorithm.

In order to illustrate the dynamic behaviour of the reactor, Figure 5 shows a dynamic profile of the reactor exit temperature for a disturbance in the coolant fluid temperature, Tr, with and without an optimization of the operation conditions. The reactor is very sensitive to changes in this operation parameter, and it has a typical asymptotic dynamic behavior, mainly for the problem without optimization.

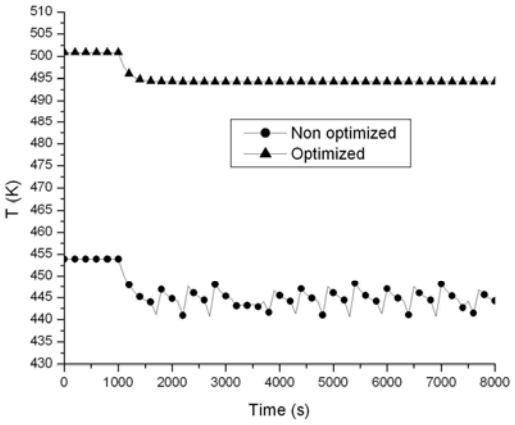

Fig. 5. Exit temperature profile of the reactor without and after the optimization.

As observed in Figure 5, the reactor shows an oscillatory dynamic behaviour for temperature profile without optimization. After the optimization, temperature goes to around 500K, however, without an oscillatory behaviour.

Analyzing the response surface of the objective function (Figure 6), the profit increases when FB and Tfo increase. It can be seen from Fig. 4 that after 10 generations, the optimal solution was obtained. The solutions show that although FB has little effect on profit, interactions between FB and Tfo are significant.

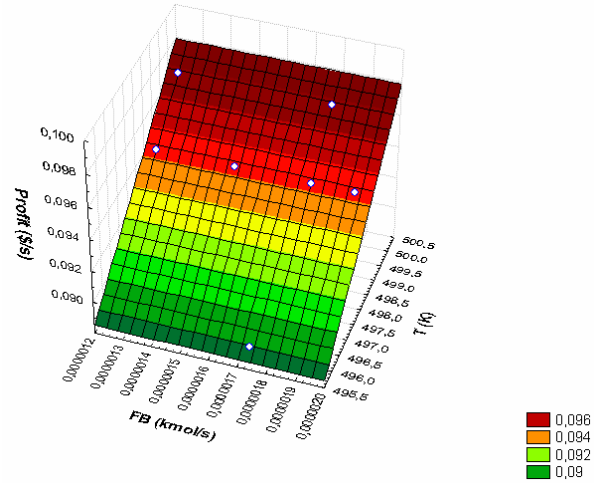

Fig. 6. Profit surface after 10 generations.

6. CONCLUSIONS

Results show that the model allowed the prediction of the main characteristics of the dynamic behaviour

of the reactor in face to changes in operational parameters. The solutions obtained using the genetic algorithm (GA) gave suitable results to optimize the process by adjusting parameters as the reactor temperature and the flow rate of reactant B. It was observed that it is possible to reach operation conditions that ensure a suitable profit and an acceptable compromise with the specifications of the desired product.

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