OPTIMIZATION STRATEGY FOR MAXIMIZING PRODUCTION OF CYCLOHEXANOL

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Abstract: The main issue objective of this work is to implement an optimization strategy through a two-layer structure where the search for optimal operational variables is made through of the algorithms optimization - Successive quadratic programming – SQP. The considered process is based on the hydrogenation of phenol for production of cyclo-hexanol in the presence of a nickel catalyst. The objective function to be considered for the optimization is to maximize production of cyclo-hexanol, through a steady-state model while maintaining the conversion of phenol at the exit of the reactor under environmental constraints. Factorial design and response surface analysis in a simulation study of a three-phase catalytic slurry reactor is used to identify the impact that each variable has on the system as well as the interaction among then. An improvement of 5.5% in cyclo-hexanol conversion was observed for the optimized variables (Q_H = 259 kg/h, Q_{Ni} = 53 kg/h).The resulted model for the response surface was used as a first objective function for the SQP optimization. The optimization results can be then used as set-point values to the advanced controller.

Keywords: hydrogenation, dynamics modeling, response surface analysis, optimization, three-phase catalytic reactor; integration of control and optimization, algorithms, simulation, steady-state modeling.

1. INTRODUCTION

Hydrogenation reactions are widely applied industrially and reactors have been designed for this purpose. It is a highly non-linear process, multivariable, with exothermic reaction. Model assessment has mostly been reported for a single reaction, or for reactions obeying simplified kinetic laws under isothermal conditions. Nevertheless, exothermic reactions undergoing a multi-step reaction scheme with complex kinetics are industrially of a main interest, and rigorous comparisons of the performances of several multiphase reactors for such reactions have hardly

ever been published (Bergault et al., 1997). With computational advances the process simulation has become an important tool for study and optimization of complex processes that normally involve a large number of variables. Hence, it is necessary to submit the process to an initial screening design prior to optimization (Kalil et al., 2000). The methodology of Placket & Burman (1946) is a tool for this initial screening, since it makes it possible to determine the influence of various factors with only a small of number of trials, instead of using more extensive factorial design, which would furnish more complete information, but which involves unfeasible complexity. If, however, the process is subjected to initial screening, response surface analysis can be limited to the most critical variables for the determination of their optimal bands of operation. Performing optimization encompasses the task of establishing process and economics models, choosing a predictive controller and modeling the optimization strategy. Yet no final answer has been given on how best to integrate the control and optimization of the operation of the reactor. Different optimization approaches have been attempted in the literature, for example, the two-layers approach where the economic optimization problem is solved separately from the control one. In this approach the economic optimization makes use of a rigorous steady-state process model and determines the set-point values for some controlled variables. Some approaches are just related to the selection of outputs and their appropriate reference values that will guarantee a probable optimal operation of the plant.

SQP algorithms (Sequential quadratic programming) are detached as non-linear (PNL) programming algorithms for the resolution of problems of no-large scale. More effective researchs have been done with the use of the SQP method, particularly for applications in complex systems. Robust algorithms have been considered and the difficulties that are found are detailed for solving the non-linear problems (Tvrzská de Gouvêa & Odloak, 1998; Ternet & Biegler, 1998; Lúcia et al., 1996). In order to reduce the convergence problems of some SQP algorithms, it is crucial to understand and identify the main difficulties of the considered optimization problem. The present work introduces response surface analysis in a simulation study of a three-phase catalytic slurry reactor to determine the operational conditions that lead to high conversion of the desired product. It is also proposed the use of statistical model from surface response analysis methodology to built of the objective function to be used in the SQP optimization algorithm.

2. MATHEMATICAL MODELING

2.1 Process modeling

The reactor considered in this work is constituted of a series of tubes, immersed in a boiler. In fact, they consist of concentrical tubes, where the reactants flow through the tubular as well as through the external annular region, while the coolant fluid flows inside the other regions. The model formulation was made focusing on the hydrogenation reaction of phenol to obtain the cyclo-hexanol (CHL), in the presence of a catalyst Ni/SiO_2 catalyst (Santana, 1999). The model consisted on mass and energy balance equations for the reactants and the coolant fluid. The scheme utilized to represent the reactor is shown in Figure 1 (Toledo et al., 2001).

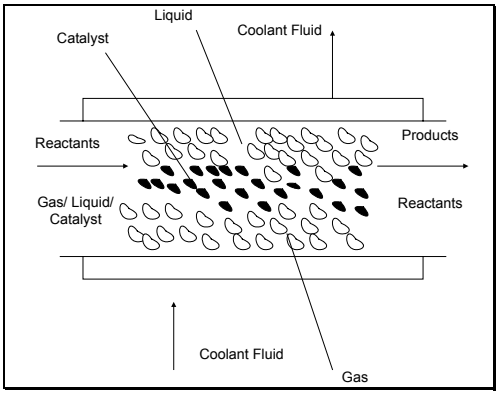

Figure 1. Three-phase reactor.

The following hypothesis were adopted when developing models that were studied in order to represent the steady-state behavior of the reactor by Santana (1999): a) plug-flow for the reactant and thermal fluid; b) homogeneous suspension (liquid + solid), considered as a pseudo-fluid; c) negligible pressure variations; d) reaction of the type: A (g) + νB (l) $\rightarrow$ νC (l), occurring at the catalyst and with a kinetic that is dependent of the concentrations of A and B; e) no phase change in the system; f) solid phase considered as pseudo-stationary. The radial dispersion has been neglected for the fluid phase, which is a very common assumption in multiphase reactor as normally found in literature. The kinetic expressions and the mass and balance equations were obtained from Gianetto & Silveston (1986), Coussemant & Jungers (1950) and Froment & Bischoff (1990). The operational parameters of the reactor, mass and energy balance coefficients, and physical properties have been considered constant. Some of these parameters are generated by correlations (Santana, 1999).

3. SEQUENTIAL QUADRATIC PROGRAMMING (SQP)

Sequential quadratic programming, or successive, or recursive, or iterative, or methods of the metric variable with restrictions, are the various form of SQP method representation, which is basically used to solve the equations of Karush-Kuhn-Tucker (KKT), or first-order conditions. The optimal result is obtained by the solution of the no-linear system of equations. The basic idea is the Newton method approach, generally used to decide problems with no restrictions. For this case study, it was necessary to establish an objective function, which is dependent upon the decision variables, and to include the restrictions related to these variables, so that the optimization problem may be previously defined. In order to reach well implemented optimization by the SQP method, an adequate choice of the decision variables must be done, with no convergence limitations. For this, it was previously used the experimental design and the response surface analysis as basis for a first optimization aiming to a definitive formulation of the SQP problem.

4. STATISTICAL ANALYSIS OF THE PROCESS

4.1Plackett-Burman Design

Packett-Burman (PB) designs are very useful for picking the most important factors from a list of candidate factors (Kalil et al., 2000), and require fewer runs than a comparable fractional design Haaland (1989). For screening purpose, ten variables on cyclo-hexanol conversion (X_{CHL}) were evaluated. The different variables were prepared in two levels, -1 for low level and +1 for high level, based on a Plackett-Burman statistical design. Dummy variables are used to estimate the standard error during analysis of data. Ten variables were considered in this study: Feed reactant temperature (T_F = 175.5 – 214.5°C), feed reactant pressure (P = 27 – 33 Kgf/cm^2), catalyst rate (Q_{Ni} = 225 – 275 kg/h), hydrogen feed rate (Q_H = 270 – 330 kg/h), coolant rate in tubes 1 to 6 (Q_{C1} = 657 – 803 kg/h, Q_{C2} = 3060 – 3740 kg/h, Q_{C3} = 5400 – 6600 kg/h, Q_{C4} = 1044 – 1276 kg/h, Q_{C5} = 405 – 495 kg/h, Q_{C6} = 234 – 286 kg/h).

4.2Response surface analysis

Once the relevant variables were selected by screening, simulations were planned to obtain a linear or a quadratic model, consisting of 2^n trials (n = number of relevant variables) plus a star configuration, with a central point. Surfaces were then built using the obtained model for the statistically significant variables. The software STATISTICA (Statsoft v. 5.0) was used to analyze the results.

5. THE OPTIMZATION STRATEGY AND THE INTEGRATION PROBLEM

The resulted model for the response surface was used as a first objective function for the SQP optimization. The optimization results can be then used as set-point values to the advanced controller. The real time process integration involving the optimization and control of the process, a step still being developed by the authors, is the final aim of the work. The process integration will be done with the two-layers approach, schematically shown in Figure 2.
In this approach, the control is set in a hierarchical structure, where an optimization layer calculates the set-points to the advanced controller, which is based on the Dynamic Matrix Control (DMC) procedure. The optimization layer is composed of an objective function and a steady-state model for the process. The control layer is composed of an economic objective function and a dynamic model for the process. Numerical results can, then, be used as comparative parameters for others strategies of the integration, such as one-layer approach for the cyclo-hexanol production.

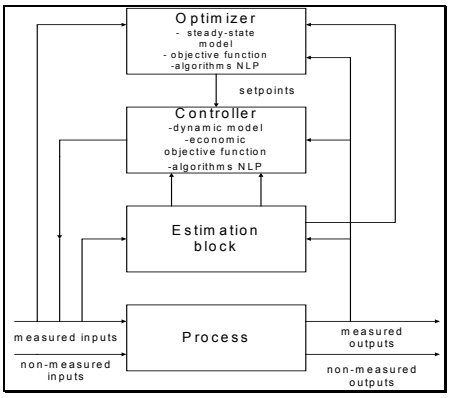

Figure 2– Two-layers Approach.

6.0 RESULTS AND DISCUSSION

6.1Evaluation of factors affecting cyclo-hexanol conversion (X_{CHL})

The experiments (simulations) were conducted according to the PB planning (which is composed by 20 trials) for the ten specified variables. The results of cyclo-hexanol conversion (X_{CHL}) were obtained for each trial. The Pareto chart was used for identifying which estimated effects are the most important. As it can be seen (Figure 3), only two parameters, catalyst and hydrogen rate (Q_{Ni} and Q_H, respectively) were statistically significant for (X_{CHL}). The negative values of effects indicate that higher values of cyclo-hexanol conversion can be obtained for lower values of Q_{Ni} and Q_H.

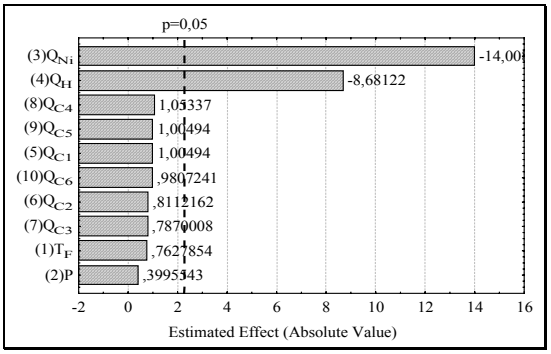

Figure 3. Pareto chart of effects for X_{CHL} (at 95% of confidence level), for the PB design.

6.2Process optimization - response surface analysis

A first central composite design (CCD) was carried out for the main variables Q_{Ni} and Q_H. It consisted on a complete factorial design 2^2 (levels –1 and +1) plus a star configuration (levels $-2^{1/2}$ and $+2^{1/2}$), with a central point (level 0). The central point is normally used with repetition for error estimation. In this case, however, only one point was used because there are no errors in computer simulations. The other eight variables were fixed at their central point of the PB design. Figure 4 shows the representation of the first central composite design results. Best values of (X_{CHL}) were obtained between simulations (trials) 1 and 6. Consequently, a new region, with lower values for Q_{Ni} and Q_H, must be explored. In such case, new simulations were carried out for values of Q_{Ni} and Q_H lower than 191.1 kg/h and 262.1 kg/h, respectively, and best value for (X_{CHL}) was obtained for Q_{Ni} = 53.0 kg/h and Q_H = 259.0 kg/h. Thus, a second CCD (similarly to the first CCD) was developed around this region for the determination of the optimal point. By the analysis of variance (ANOVA) for X_{CHL}, the F-test showed that the model was reliable since the calculated F-value was 4.29 times greater than the listed F-value, for 90% of confidence level. As a practical rule, a model has statistical significance when the calculated F-value is at least 3-5 times greater than the listed value (Kalil et al., 2000). The response has a correlation coefficient of 0.9705.
The model, expressed by equation (1), was generated by non-linear multiple regression of the data and is a function of the more significant variables,

$$X_{CHL} = 0.9245 - 0.001 \cdot Q_{Ni} - 0.0028 Q_{Ni}^2 + 0.0018 Q_H - 0.0024 Q_H^2 - 0.0011 Q_{Ni} \cdot Q_H \quad (1)$$

where Q_{Ni} and Q_H are in coded form. The response surface is represented in Figure 5. As it can be seen, X_{CHL} reaches a maximum value for Q_{Ni} = 53.0 kg/h and Q_H = 259.0 kg/h (corresponding to the central point of the second CCD).

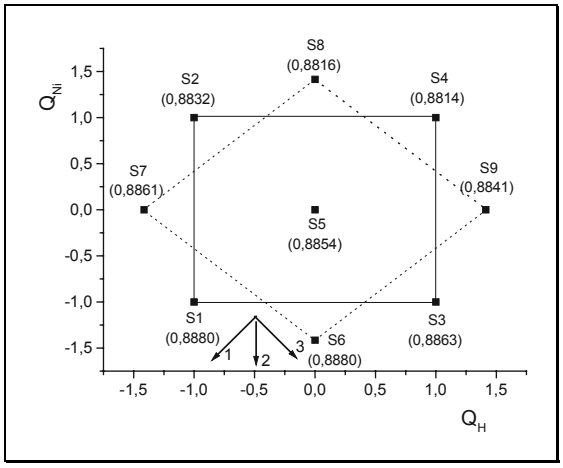

Figure 4. Representation of first central composite design. The values in brackets are the X_{CHL} results of simulations (S). The arrows 1, 2 and 3 indicate the trajectory at which Q_{Ni} and Q_H must be investigated (between S1 and S6) for the obtainment of higher X_{CHL} values.

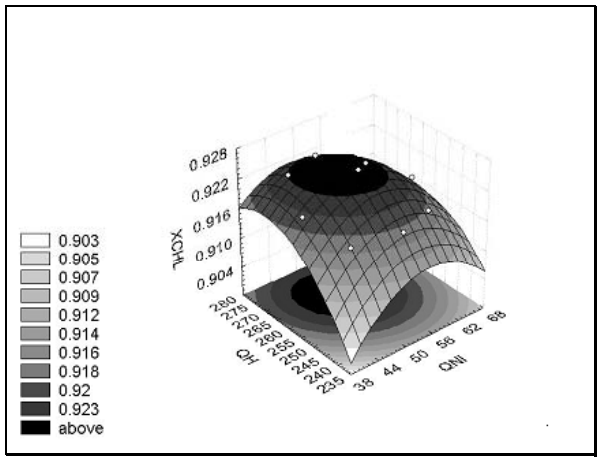

Figure 5. Response surface of conversion (X_{CHL}) expressed as a function of Q_{Ni} and Q_H, obtained for the second central composite design. Q_{Ni} e Q_H with real values.

6.3 Results of SQP Optimization

The following equation was used as objective function in the SQP implementation and represents cyclo-hexanol conversion dependency from the decision variables Q_{Ni} and Q_H. This equation was obtained from the response surface analysis where Q_{Ni} and Q_H are in real form and aims to cyclo-hexanol conversion maximization and, consequently, cycle-hexane minimization (undesired byproduct).

$$X_{CHL} = 0.9245 - 0.001 \cdot \left(\frac{(Q_{Ni} - 53.0)}{(8.5)}\right) - 0.0028 \cdot \left(\frac{(Q_{Ni} - 53.0)}{(8.5)}\right)^2 + 0.0018 \cdot \left(\frac{Q_H - 259.0}{12.4}\right) - 0.0024 \cdot \left(\frac{Q_H - 259.0}{12.4}\right)^2 - 0.0011 \cdot \left(\frac{(Q_{Ni} - 53.0)}{(8.5)}\right) \cdot \left(\frac{Q_H - 259.0}{12.4}\right) \quad (2)$$

The advantage of using simplified process models in optimization problems is the reduction in convergence problems, so that the optimum point is reached easier.

The SQP code was implemented according to the algorithm developed by Schittowski (1995). Simultaneously, it was tested the routines of optimization of Martinez et al (2000), and the BDCONF routine (from IMSL library).

The optimal values of Q_{Ni} and Q_H were obtained from the routine EASY of Martinez et al. (2000) and were close to the statistical results (second CCD). The maximization of the objective function was reached at a CPU time of 1.5625 10-0.002 seconds. In a second stage, the original steady-state model of the process was incorporated in the routine following pre-adjusted parameters and convergence was obtained under a rational computational time. The convergence was possible through a strait band of nickel and hydrogen rates and the optimal point (X_{CHL} = 0.9245) was obtained for Q_{Ni} = 53.0 kg/h and Q_H = 259.0 kg/h.

The routines of the IMSL Fortran's library were tested and convergence problems were observed due to the high dimension of the problem. The DNCONF subroutine includes searching in unfeasible regions resulting in convergence limitations for the optimization problem.

7. CONCLUSIONS

In this paper a procedure for real time optimization was proposed through the coupling of SQP based algorithm and statistical analysis. The optimization results will be then used as set-point values to the advanced controller. Experimental design techniques was used to identify the main process variables with a significant impact on the process behavior, after a just screening by PB methods. Response surface analysis were implemented to find out a possible optimal region as well a to generate a reduced model statically validated.

Response surface methodology was found to be a powerful tool for the optimization of complex processes such as three-phase catalytic slurry reactor. In this study, an improvement of 5.5% was achieved for cyclo-hexanol conversion (0.9245 at the second CCD against 0.8763 at the previous PB design), with a decreasing in the by-product conversion. Plackett-Burman design offers a good and fast screening procedure and mathematically computes the significance of a large number of factors in one experiment, which is time saving and maintains convincing information on each component. The rate of hydrogen (Q_H) and catalyst (Q_{Ni}) showed strong effect on the cyclo-hexanol production and consequently should have special attention in the designing and operating process control. Surface response methodology is an interesting procedure to be used as alternative optimization or even to identify the feasible region for optimal operating conditions. In this case, the model obtained at the surface response methodology (eq. 1) was used as a first objective function for the SQP optimization..

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