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Identification Experiments for the Optimizing Control of Multiple Recycle Processes

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Abstract: An identification experiment procedure for the optimizing control of reactor/separator processes with multiple material recycle streams is proposed. Steady state relation between the cost related variables and the operation variables are approximated through response surface models. To minimize the perturbation due to the identification experiments, Latin hypercube design is applied to the selection of the sampling points and their implementation sequence is determined by the application of the traveling salesman problem. The procedure is applied to the simulated HDA (hydrodealkylation of toluene) plant to identify an unconstrained optimal operating condition.

Keywords: Plantwide control, recycle system, optimization, identification, response surface methodology, traveling salesman problem

1. INTRODUCTION

Material recycles are common in chemical processes. When a reactor is so designed that the per-pass conversion is well below 100%, separators are introduced to recover and recycle the unconsumed reactants. Often, a reactor per-pass conversion is designed at a moderate value due to increased side reactions and increased reactor construction costs for a higher conversion. For many chemical plants, the most important trade-off for the optimization of both flowsheet design and operating policy is between selectivity losses at high reactant conversion and recycle costs at low reactant conversion (Ward et al., 2004).

The simplest configuration of the reactor-separator-recycle (RSR) system which comprises a CSTR and distillation column process with one material recycle has been extensively studied as one of the representative examples of plantwide control problems (Papadourakis, 1987; Luyben, 1994; Wu et al., 1996; Larsson et al., 2003; Seki and Naka, 2008), since the existence of even a single material recycle complicates the overall behavior of the combined units. Larsson et al. (2003) has shown that the optimal operation of such a system is sometimes unconstrained due to the trade-off between the reaction yield and the separation cost. When the optimal operation does not reside on process constraints, extra effort becomes necessary to search for the exact position of the optimal operating point.

When more than one reactants are involved, RSR systems may include multiple recycles. Processes where both gas and liquid are recycled as shown in Fig. 1 fall into this category, so that multiple recycle processes are not rare. With multiple recycle systems, there are at least the same number of operational degrees of freedom as the number of the material recycle streams (Ward et al., 2004), so that the search for optimal operation becomes much more complicated.

To find optimal operation, the most rigorous approach would be to use a first principles model and solve a constrained nonlinear optimization problem. Although such an approach, known as RTO (Real-Time Optimization), has been widely adopted in the industry (Cutler and Perry, 1983), it demands large effort to develop and maintain the model and the computational load is quite high.

Instead of resorting to first principles nonlinear process models, experimentally identified empirical process models may be utilized. One popular empirical nonlinear modelling method is a Response Surface Methodology (RSM) (Myers et al., 2009). In the RSM, a low-order (typically first or second order) polynomial is used to describe the relation between the target variable and independent variables. Once the RSM models are obtained, constrained nonlinear optimization can be applied on-line to determine optimal operation. With the RSM, efforts for

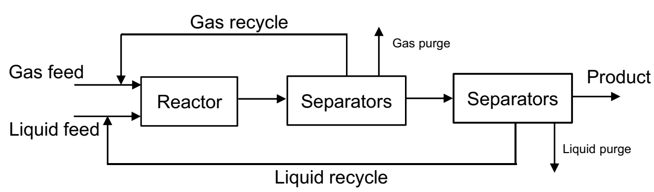

Fig. 1. Reactor/separator system with gas and liquid recycle streams

model development could be reduced compared with the rigorous first principles modelling approach.

One of the important aspects of applying such empirical approach is to keep the identification experiment from disturbing the process too much. In this paper, we introduce an identification experiment procedure, which minimizes perturbation to the plant, for finding the optimal operating point of reactor/separator processes with multiple material recycles. As a well-known example of chemical plants with multiple recycles, the HDA (hydrodealkylation of toluene) process (Douglas, 1988) is studied. This process is considered to be an important test-bed problem for design of new control structures and has been exhaustively studied by many researchers (Ng and Stephanopoulos, 1996; Luyben et al., 1997; Luyben, 2002; Qiu et al., 2003; Konda et al., 2005; Araujo et al., 2007a,b).

This paper is organized as follows. In the next section, the proposed identification procedures are introduced. In section three, the identification experiments are demonstrated on the HDA process by using the dynamic simulator UniSim. Finally, concluding remarks are made.

2. PROPOSED METHOD OF IDENTIFICATION EXPERIMENTS

2.1 Response Surface Methodology and Latin Hypercube Sampling

We assume that the operation cost c is described by

$$c = \sum_i c_i y_i$$

where y_i is a cost related process variable such as feed rates, compressor power inputs, distillation column energy inputs, etc., and c_i is the price of each process variable.

In the Response Surface Methodology (RSM), the steady state relation between the process variable y_i and independent variables u_k, $(k = 1, \cdots, M)$ is approximated by a low order polynomial. In this study, a second-order polynomial:

$$\hat{y}_i = \sum_k \alpha_{i,k} u_k^2 + \sum_{j<k} \beta_{i,j,k} u_j u_k + \sum_k \gamma_{i,k} u_k + \delta_i, \quad (1)$$

is used, where $\hat{y}_i$ is the approximation of y_i.

Once the response surface models for the process variables y_i are obtained, the cost approximation $\hat{c}$ can be made by

$$\hat{c} = \sum_i c_i \hat{y}_i. \quad (2)$$

Then, nonlinear optimization calculations may be applied to (2) with u_k as decision variables. When constraints on some of the variables have to be considered, response surface models for those variables are incorporated in the optimization problem. It can be expected that the computation load is considerably reduced, compared with the approach using a rigorous first principles model.

Identification experiments will be performed to obtain data at several sampling points and a least squares fit will be applied to determine the coefficients $\alpha_{i,k}, \beta_{i,j,k}, \gamma_{i,k}$, and δ_i in Eq. (1). For obtaining an accurate response surface model, it would be desirable to collect as many sampling points as possible, which may be chosen to have a uniform random distribution in the search range.

On the contrary, from the viewpoint of plant operations, the number of sampling points should be made as small as possible to keep the perturbation to operating plants acceptably small. To reduce the number of sampling points while retaining the accuracy of the model, Latin hypercube sampling (LHS) can be readily applied (McKay et al., 1979). The LHS is a form of stratified sampling, which is commonly used to reduce the number necessary for a Monte Carlo simulation to achieve a reasonably accurate random distribution.

The sampling points thus determined are defined as

$$S_i = (u_1^i, u_2^i, \cdots, u_M^i), \quad i = 1, \cdots, N \quad (3)$$

where N is the number of the sampling points.

2.2 Application of Traveling Salesman Problem

The problem now is "in which sequence the LHS points (3) should be implemented?" From the plant operation point of view, it is desirable to keep the process disturbance as small as possible. It is proposed here that the sequence is determined to minimize the total "distance" required to implement all the samples; the distance between the sample points i and j is defined simply by:

$$d_{i,j} = \sqrt{\sum_k (\frac{u_k^i - u_k^j}{s_k})^2},$$

where s_k is a scaling factor for the independent variable k. This is equivalent to solving the well-known traveling salesman problem (TSP): Given a set of cities along with the cost of travel between each pair of them, what is the cheapest way of visiting all the cities and returning to the starting point? (Applegate et al., 2007) Even though the problem is computationally difficult to solve, a large number of heuristics exist, so that it is easy to get approximate solutions by using optimization technique such as genetic algorithm.

3. APPLICATION TO THE HDA PROCESS

3.1 HDA Process

Process description The HDA process is a petrochemical process for producing benzene through hydrodealkylation of toluene. Figure 2 shows the process flow diagram. The process consists of a plug flow reactor and separators with gas and liquid recycle streams.

Fresh toluene and hydrogen (97% hydrogen and 3% methane) are introduced to the process and they are mixed with the recycled liquid and gas streams. This reactant mixture is pre-heated by the feed-effluent heat exchanger (FEHE) and then heated up to the reaction temperature by the furnace before being fed to the adiabatic PFR. Two main reactions take place inside the reactor:

Main reaction:
$C_6H_5CH_3 + H_2 \longrightarrow C_6H_6 + CH_4$

Side reaction:
$2C_6H_6 \longleftrightarrow C_{12}H_{10} + H_2$

The reactor effluent is quenched with a portion of the liquid stream from the gas/liquid separator and further cooled by the FEHE and the cooler before being fed to the gas-liquid separator. A part of unconverted hydrogen and methane in the overhead vapor from the separator is purged while the remainder is recycled to the reactor. The stabilizer column processes the liquid from the separator to remove remaining hydrogen and methane as the overhead product. Benzene is recovered from the stabilizer liquid outlet as the desired product at the product column top. Finally, in the recycle column, toluene is separated from diphenyl, as the distillate, and recycled back to the reactor. The three columns are equipped with reboilers at the bottom and condensers and reflux drums at the top, although they are not shown in the figure.

The model of the HDA process used in this study is a modified version of the model by Luyben (2002) and it has been implemented on the commercial dynamic simulator UniSim. It should be pointed out that the reaction kinetics used in this study is different from those used in Luyben (2002) and Araujo et al. (2007a), which do not account for the selectivity loss at a higher conversion. The activation energies and related kinetic parameters are modified in this study, so that the selectivity loss may become significant at a higher conversion, as discussed in Douglas (1988) and Phimister et al. (1999).

Regulatory control configuration Figure 2 also shows the regulatory multi-loop controller configuration. The pressure controller, which manages the gas components holdup and regulates the system pressure, introduces the fresh H_2 gas as the controller handle. The hydrogen mass balance, consequently the methane balance, is handled by the composition controller on the gas recycle stream, which manipulates the purge flow rate. The total gas flow rate, that is the sum of the fresh gas and recycle gas, is controlled by the compressor to keep the hydrogen/aroma concentration ratio above the prescribed value of 5.0.

The reaction is controlled by regulating the inlet temperature with the furnace fuel input as the controller handle. The temperature of the reactor effluent is controlled by the flow rate of the quench stream from the gas/liquid separator bottom, which is kept at 621°C.

In the stabilizer, the gas purged from the top is used for the pressure control. The benzene leak from the top is regulated by the composition controller whose handle is the condenser energy input at the reflux. Also, at the bottom, composition control is implemented to keep the methane concentration below a certain value by the reboiler.

In the product and recycle columns, the composition controllers are implemented for the top and bottom streams with the reflux flow rates and reboiler boil-ups as the controller handles, respectively. In these columns, the pressures are regulated by condenser heat removals.

From the top of the recycle column, almost pure toluene is recycled to the liquid feed tank. The fresh make-up toluene is introduced to the feed tank as the level controller handle. The effluent from the feed tank is put on flow control. This configuration follows the Luyben's rule(Luyben, 1994; Bildea and Dimian, 2003)

The throughput can be adjusted by the total liquid feed rate and the toluene conversion. The toluene conversion is regulated by manipulating the setpoint of the reactor inlet temperature controller.

3.2 Identification Experiments for Optimizing Control

Araujo et al. (2007a) found that the following process variables are likely to be constrained with the optimal operation:

Gas loop pressure	-	upper limit
H_2/aroma ratio at the reactor inlet	-	lower limit
Quench temperature	-	upper limit

They also showed that the current column design make the energy consumption of the columns insensitive to the composition controller settings.

Then, for the optimizing control of the HDA process, the remaining three operational degrees-of-freedom are chosen as the toluene conversion, the H_2 concentration of the gas recycle stream, and the total liquid feed rate. In fact, the first two variables were used as the key design parameters at the original process design phase (Douglas, 1988), which may give intuition about the inherent trade-off in the optimal operation policy.

Example: the maximum throughput case It is assumed that the total liquid flow rate is constrained at its maximum, aiming at maximizing the production rate: the setpoint to the total liquid flow rate controller is held constant at 145kmol/h.

The setpoint to the toluene composition controller x_{TOL} is varied in the range of 0.05 - 0.2, while the setpoint to the H_2 composition controller on the gas recycle $y_{\mathrm{H_2}}$ is changed in the range of 0.25 - 0.35. On the basis of the LHS, the total of 30 sample points are generated and their implementation sequence is determined through the application of the TSP. For the LHS, the MATLAB code available in the public domain (Minasny, 2004) is used in this study. We assume that the process is initially at $x_{\mathrm{TOL}} = 0.13$ and $y_{\mathrm{H_2}} = 0.3$, and the sampling starts at this point and comes back to the same operating point after scanning the 30 samples. For the determination of the sampling sequence, the MATLAB code by Kirk (2011) for solving the travelling salesmen problem is utilized. Figure 3 shows the obtained sampling sequence.

The sequence is implemented on the simulated HDA plant. When the new sample point is implemented, if the move from the previous sample happens to be too large, the move is divided into several uniform steps so that a single step becomes smaller than a prescribed limit. Such steps are implemented every 1 hour. After reaching the new target value, the setpoints are held constant for 5 hours to wait for the process to reach steady states.

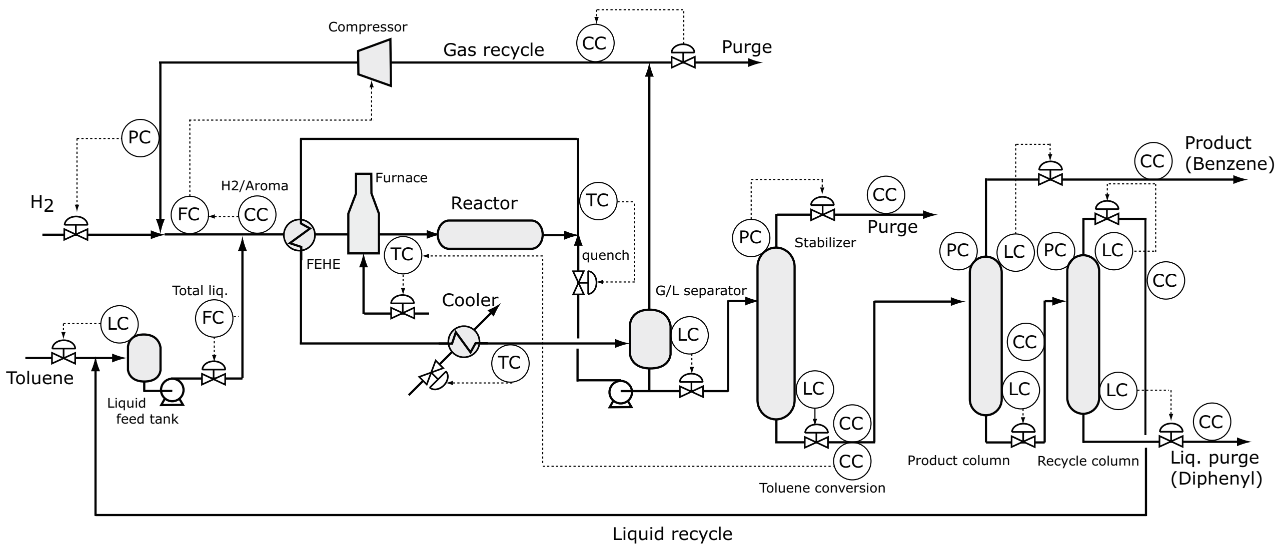

Fig. 2. The HDA process flow diagram with regulatory control loops

Figure 4 shows the time evolution of the two independent variables in the identification experiment. For the calculation of the response surfaces, the time average data from the time period which is just before the next new move are employed.

Figure 5 and 6 show an example of the obtained response surface models for the benzene and diphenyl production. Decreasing the toluene composition, equivalently increasing the reactor temperature for the higher toluene conversion, increases the benzene production but also increases the by-product diphenyl, resulting in the lower selectivity. This is one of the major trade-off concerning the economically optimal operation in this process.

Combining the obtained response surface models, the operation cost in terms of the two operational degrees of freedom, that is x_{TOL} and y_{H_2}, are constructed. Table 1 shows the list of process variables related to the cost calculation and their prices. Figure 7 shows the response surface for the operational cost. The figure implies that the optimal operation is unconstrained. When the prices change, the model for the operation cost can be easily reconstructed by using Eq. (2), without performing additional experiments.

Table 1. Variables associated with cost calculation and their prices

Stream name	price	unit
Fresh toluene feed	24.0	\$/kmol
Fresh gas feed	2.9	\$/kmol
Fuel	1.9×10^{-6}	\$/kJ
Steam	2.22×10^{-8}	\$/kJ
Cooling water	1.77×10^{-5}	\$/kJ
Electricity	1.11×10^{-5}	\$/kW
Benzene product	-39.9*	\$/kmol
Diphenyl(fuel)	-11.9	\$/kmol
Toluene(fuel)	-7.4	\$/kmol
Benzene(fuel)	-6.2	\$/kmol
Methane(fuel)	-1.7	\$/kmol
Hydrogen(fuel)	-0.54	\$/kmol

*Negative values imply profit.

Figure 8 compares the response surface models for the cost approximated by the 30 sampling points and 100 sampling points. There is some discrepancy in the optimal operating conditions predicted by the two models, as shown in

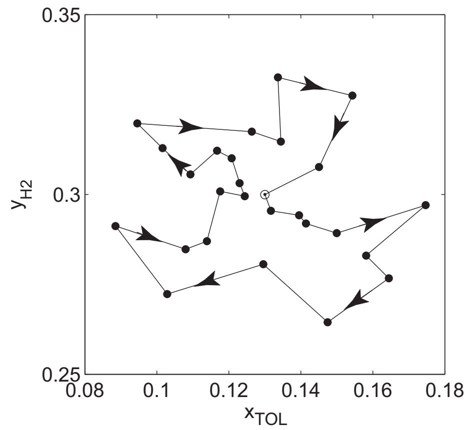

Fig. 3. Sampling sequence obtained by solving the traveling salesman problem. ∘: start and end sampling point.

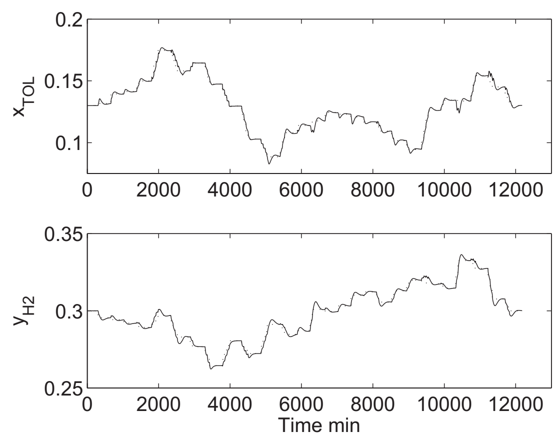

Fig. 4. Time plots of the independent variables. Dotted: setpoint, solid: actual.

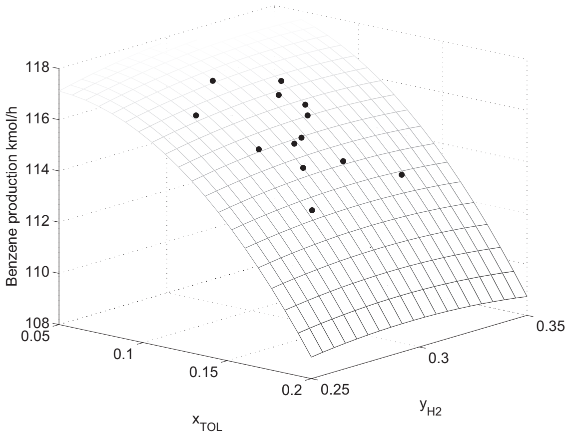

Fig. 5. Response surface for the benzene production. The dots are the sampling points.

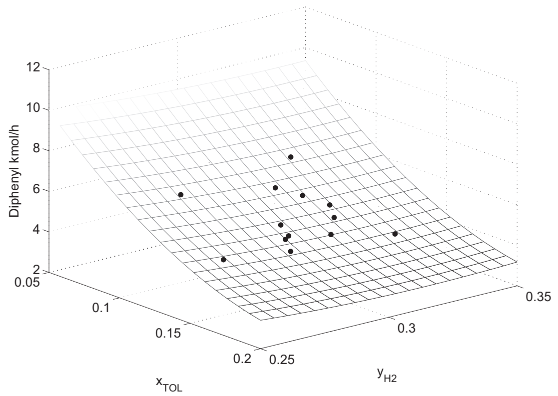

Fig. 6. Response surface for the diphenyl production. The dots are the sampling points.

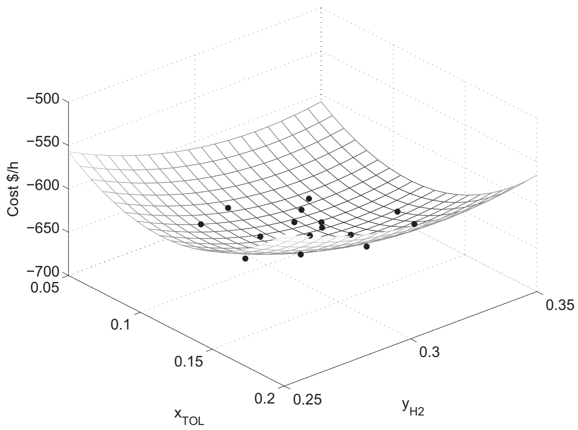

Fig. 7. Response surface model for the operation cost

Table 2. However, the optimization landscape is relatively flat, so that the associated cost is almost the same.

4. CONCLUDING REMARKS

In implementing such empirical approaches as proposed in this paper, it is important to have some intuition about the nature of the optimization problem: What are the dominant trade-offs? How does the optimal operating point change as constraints, disturbances, and prices change? For this purpose, off-line optimization study using first principles model is considered equally important. That would help define and reduce the number of independent variables and narrow the the search range. If the optimal operating point is found to lie always on a certain set of constraints, it would be no longer necessary to implement an RTO system. If the optimization problem is found to be nonconvex, application of this approach would be unlikely, because higher order polynomial approximation of complicated response surfaces would be very difficult.

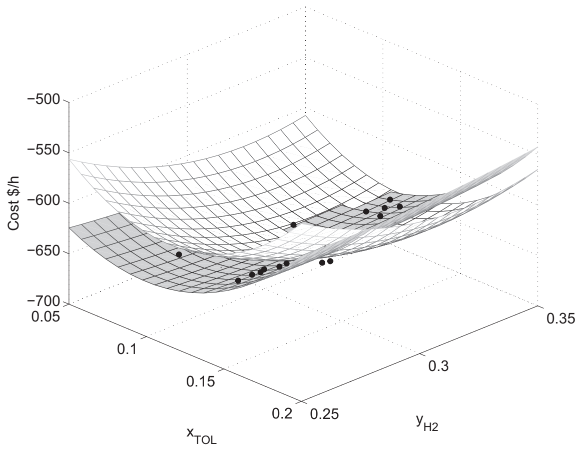

Fig. 8. Comparison of the cost models obtained from data with different numbers of sampling points. Shaded surface: 100 sampling points. Non-shaded surface: 30 sampling points.

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Table 2. Comparison of optimal operations predicted by the two response surface models

No of sampling points	x_{TOL}^{opt}	$y_{\mathrm{H}_2}^{opt}$	Cost \$/h
30	0.161	0.292	-653
100	0.145	0.276	-652

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