Measurement-based Run-to-run Optimization of a Batch Reaction-distillation System

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Abstract

Measurement-based optimization schemes have been developed to deal with uncertainty and process variations. One of the methods therein, labeled NCO tracking, relies on appropriate parameterization of the input profiles and adjusts the corresponding input parameters using measurements so as to satisfy the necessary conditions of optimality (NCO). The applicability of NCO-tracking schemes has been demonstrated on several academic-size examples. The goal of this paper is to show that it can be applied with similar ease to more complex real-life systems. Run-to-run optimization of a batch reaction-separation system with propylene glycol is used for illustration.

Keywords: Dynamic optimization, Reaction-distillation system, Batch processes, Measurement-based optimization, Run-to-run optimization.

1. Introduction

The sequence of reaction and separation steps represents a process configuration that is frequently encountered in batch chemical processing (Schenk et al., 1999). Optimization of such processes falls under dynamic optimization, where a given performance index is minimized while satisfying path and terminal constraints. In the presence of uncertainty (modeling errors, disturbances), the constraints are typically satisfied by applying a conservative policy that is non-optimal in most cases (Terwiesch et al., 1994).

One possibility of reducing this conservatism consists of using measurements to improve the performance of the real process. This can be accomplished via model refinement and re-optimization (*explicit optimization*) (Eaton et al., 1990) or by adapting the inputs directly (*implicit optimization*) (Srinivasan et al., 2003). This paper considers the use of measurements for optimization via the tracking of the necessary conditions of optimality (NCO) (Srinivasan et al., 2003). NCO tracking attempts to meet the NCO by adjusting the parameters of a solution model that is typically generated from numerical optimization of a nominal (tendency) model. The optimal inputs are typically discontinuous, but are continuous and differentiable within each arc. These inputs are dissected into various parts and appropriately parameterized. The

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resulting free variables are then linked to the various elements of the NCO, and appropriate adaptation laws are provided.

So far, NCO tracking has been investigated via simulation of academic-size problems involving relatively low-order single-input systems (François et al., 2002, Srinivasan et al., 2003). However, the scheme should also be applicable to more complex systems since the ease with which it is implemented does not depend on the complexity of the system (in terms of the number of states or equations), but rather on the possibility of approximating the optimal solution with a solution model. Along these lines, this paper investigates a realistic two-input batch reaction-separation system for which a large-scale rigorous model is available (337 states and about 2000 algebraic equations).

The paper is organized as follows. Section 2 describes the NCO-tracking scheme for run-to-run optimization. The problem of optimizing the production of propylene glycol is formulated in Section 3, while Section 4 generates the corresponding solution model. The optimization results via run-to-run adaptation are presented in Section 5, and Section 6 concludes the paper.

2. Measurement-based Run-to-run Optimization

The following terminal-cost dynamic optimization problem is considered:

$$\min_{u(t),t_f} \quad \phi(x(t_f),t_f) \tag{1}$$

$$\text{s.t.} \quad \dot{x} = F(x,u) \qquad x(0) = x_o$$

$$S(x,u) \leq 0 \qquad T(x(t_f)) \leq 0$$

where ϕ is the scalar cost function, x the n-dimensional states with the initial conditions x_0, u the m-dimensional inputs, and t_f the final time. F are the equations describing the system dynamics, S the ζ-dimensional path constraints, and T the τ-dimensional terminal constraints. Without loss of generality, all terminal constraints are assumed to be active in the optimal solution, the non-active ones being simply discarded.

Since optimality requires meeting the NCO, the optimization problem can be treated as a control problem via NCO tracking. The NCO consist of several parts that deal with both constraints and sensitivities. One way of enforcing the NCO is to parameterize the inputs using time functions and scalars and assign them to the various NCO parts. This assignment corresponds to choosing the *solution model* (Srinivasan et al., 2004).

In this work, the inputs are parameterized using the n_π-dimensional parameter vector π. Note that $n_\pi \geq \tau$ so as to be able to meet all terminal constraints. Also, the path constraints are assumed to be implicitly satisfied with the chosen parameterization. Then, using the fact that $x(t_f) = X(\pi)$, the dynamic optimization problem can be recast as the following static optimization problem.

$$\min_{\pi} \quad \Phi(\pi) \qquad \text{s.t.} \qquad T(\pi) = 0 \tag{2}$$

For the above static optimization problem, the constraint and the sensitivity parts of the NCO are given by (Srinivasan et al., 2003):

$$T = 0, \qquad \frac{\partial \Phi}{\partial \pi} + \nu^T \frac{\partial T}{\partial \pi} = 0 \tag{3}$$

where ν are the τ-dimensional Lagrange multipliers for the terminal constraints.

Since batch processes are intended to be run repeatedly, it is natural to exploit this feature for process optimization. This way, optimal operation can be found iteratively over several runs. Furthermore, since there is often more to gain by keeping the constraints active compared to pushing the sensitivities to zero, this study will focus on a run-to-run controller to keep the terminal constraints $T=0$ active. A gain matrix relating the input parameters to the terminal constraints (local sensitivities) is used for this purpose. Consider the $\tau \times n_\pi$ gain matrix $G = \partial T / \partial \pi$ with $n_\pi \geq \tau$ and rank$(G) = \tau$. The pseudo-inverse of this matrix can be used for decoupling (François et al., 2002):

$$\pi(k+1) = \pi(k) - G^+ K\, T(k) \tag{4}$$

where k is the batch index and K a diagonal gain matrix of dimension $\tau \times \tau$, and the superscript $+$ is used for the pseudo-inverse.

3. Batch Production of Propylene Glycol

3.1 Reaction-distillation System
The production of propylene glycol (PG) by acid-catalyzed hydration of propylene oxide (PO) is considered. In addition to the monoglycol, dipropylene glycol (DPG) and tripropylene glycol (TPG) are obtained in smaller amounts as by-products, according to the following reaction scheme:

$$PO + H_2O \rightarrow PG, \qquad PO + PG \rightarrow DPG, \qquad PO + DPG \rightarrow TPG \tag{5}$$

The three reactions are highly exothermic. A high H_2O/PO initial molar ratio favors the production of PG. Methanol is used as a solvent to break the partial solubility between water and PO. The initial conditions described by Furusawa et al. (1969) are used. The larger activation energies of the higher-order glycol reactions indicate that higher temperatures will favor the production of DPG and TPG over PG.

The reaction is carried out in a jacketed stirred tank reactor. The reactor temperature is controlled by a PID controller that adjusts the jacket inlet temperature. After the reaction stage, water and methanol are removed from the glycol mixture by distillation. The column is represented by six theoretical stages with constant volume hold up. Thermodynamic properties and liquid-vapor equilibrium are rigorously calculated using the software BibPhy32 of Prosim. The integration of the set of nonlinear differential equations is carried out by DISCo (Sargousse et al., 1999), which can handle discontinuities in the differential equations. A rigorous mathematical model of order 337 is used (see Elgue (2002) for detailed description).

3.2 Optimization Problem
Two manipulated variables are considered, the reactor temperature T_r, and the internal reflux ratio r. The objective is to minimize the operation time t_f while meeting terminal constraints on the reaction selectivity, and the yield and final mole fraction of PG:

$$\min_{T_r(t), r(t), t_f} \quad J = t_f \tag{6}$$

$$\text{s.t.} \quad \text{DAE system} \qquad 0 \leq r(t) \leq 1 \qquad T_{j,in}(t) \leq 170\ ^\circ\text{C}$$

$$y_{PG}(t_f) \geq 0.89 \qquad w_{bp}(t_f) \leq 0.04 \qquad x_{PG}(t_f) \geq 0.8$$

$$y_{PG} = \frac{n_{PG}}{n_{PO}^{o}} \qquad w_{bp} = \frac{w_{DPG} + w_{TPG}}{w_{PG} + w_{DPG} + w_{TPG}}$$

where x_{PG} is the molar fraction of PG in the reactor, y_{PG} the PG yield, n_{PG} the number of moles of PG in the reactor, n_{PO}^{o} the initial number of moles of PO in the reactor, w_i the mass fraction of glycol i in the reactor and $T_{j,in}$ the jacket inlet temperature.

4. Formulation of the Solution Model

Generating the solution model consists of (i) identifying the input arcs and choosing an appropriate parameterization, and (ii) linking the input parameters to the NCO (in this case the active terminal constraints). It can be seen from the numerical solution presented in Elgue (2002) that the optimal solution consists of three arcs: (A) the reaction phase, (B) the start up phase of the distillation, and (C) the distillation phase. All terminal constraints are active at the optimum. The reactor temperature exhibits a sensitivity-seeking arc during phase (A), which expresses the compromise between speeding up the main reaction and producing side products. In the other two phases, the reactor temperature is determined from the maximum jacket inlet temperature. As far as the internal reflux ratio is concerned, there is full reflux during phases (A) and (B) and a sensitivity-seeking arc during the distillation phase, which represents a compromise between quality and quantity in distillation.

Though different parameterizations are possible, the two sensitivity-seeking arcs are approximated by exponential functions with two parameters:

$$T_r(t) = \begin{cases} \alpha_1 + e^{\alpha_2 t} & 0 \leq t \leq t_h \\ \mathcal{T}'(T_{j,in}^{\max}) & t_h < t \leq t_f \end{cases} \qquad r(t) = \begin{cases} 1 & 0 \leq t \leq t_p \\ \beta_1 + e^{\beta_2(t-t_p)} & t_p < t \leq t_f \end{cases} \qquad (7)$$

where the operator $\mathcal{T}(T_{j,in})$ represents the reactor temperature that results from a given choice of $T_{j,in}$. The optimal solution, computed numerically, is presented in Fig. 1.
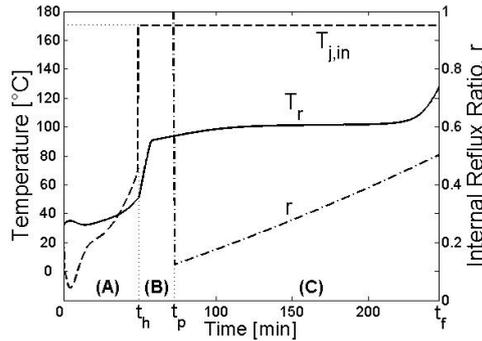

Figure 1. Optimal profiles: Reactor temperature T_r (solid), jacket inlet temperature $T_{j,in}$ (dashed) and internal reflux ratio r (dot-dashed)

Table 1. Optimal input parameters, switching times and constraints.

α_1	α_2	β_1	β_2	t_h[min]	t_p[min]	t_f[min]	x_{PG}	y_{PG}	w_{bp}
32.4	1.04×10^{-3}	-0.92	3.78×10^{-5}	49.25	73.0	247.1	0.8043	0.890	0.040

The duration of the start-up phase can be fixed a priori. Furthermore, the final time t_f can be determined on-line by stopping the distillation at a pre-determined reactor temperature that is related to the desired final concentration of PG. This way, the constraint regarding the final mole fraction of PG in the reactor is kept active in every batch. The distillation is stopped when the boiling mixture in the reactor reaches 128°C. Finally, if all PO has reacted when time t_h is reached, any further increase of t_h has no effect on the reactor concentrations. Hence, instead of adapting t_h, the reaction phase is stopped when the reactor temperature reaches some heuristically-determined value. This temperature can be chosen within a wide range with little effect on t_f. Here, it is chosen as 54°C. With these simplifications, the only parameters that need to be adapted are α_1, α_2, β_1 and β_2. The numerical optimal solution is presented in Table 1. Had the switching times t_h, t_p, and t_f been included in the parameterization, the gain in performance (reduction of t_f) would have been of only 0.5 min, i.e. 0.2%.

5. Optimization via Run-to-run Adaptation

5.1 Conservative Starting Points
The numerical optimization presented in the previous section uses a model, labeled "simulated reality", that is normally unknown. Thus, one has to have initial guesses for the parameters. Two starting points are considered to evaluate the run-to-run adaptation. The first point (Case I) comes from the numerical optimization of a conservative mechanistic model. In this model, conservatism with respect to the rate of reaction, the heat transfer efficiency and the separation efficiency of the distillation column is introduced. The second starting point (Case II) considers a guess that can be considered as an industrially-relevant solution due to low temperatures in the reaction phase and high reflux during the distillation phase. The choices are summed up in Table 2.

Table 2. Initial input parameters, initial final times and adaptation results. Improvement is computed with respect to the initial guess and loss with respect to the ideal optimal solution.

	Starting conditions					Adaptation results		
	α_1	α_2	β_1	β_2	t_f[min]	t_f[min]	Improvement	Loss
Case I	32.95	9.4×10^{-4}	-0.70	2.11×10^{-5}	298.5	255.0	14.6%	3.2%
Case II	27.0	8.6×10^{-4}	-0.85	3.9×10^{-5}	311.3	250.6	19.5%	1.4%

5.2 Adaptation Results
The measurements of the final mole fractions are considered to exhibit 0.5% zero-mean Gaussian noise. The calculation of n_{PG} at final time requires the measurement of the final product density, which is considered to have 0.2% zero-mean Gaussian noise. The gain matrix G is determined at the starting point I and the adaptation law is determined from (4). Fig. 2 shows the cost function obtained for 10 runs for Cases I and II.

In order not to violate the constraints, the margins of 0.2 and 0.01 were used for the PG yield and the fraction of higher glycols, respectively. The region where the adaptation is within the noise level is reached after 2-4 batches and the adaptation may be stopped at

this point. The column "improvement" is computed with respect to the initial guess and the "loss" with respect to the ideal optimal solution. The adaptation starting from the second point (Case II) performs better since the directions that are not adapted are closer to the optimal ones in Case II than in Case I.
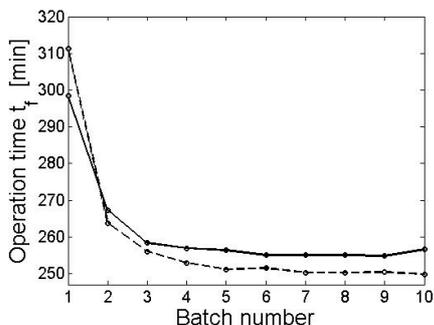

Figure 2. Evolution of final batch time. Case I: solid, Case II: dashed.

6. Conclusion

This work has considered a fairly complex reaction-separation process and showed that a simple solution model with a few adjustable parameters can be used efficiently for optimization purposes. The input parameters are adjusted using on-line and off-line measurements to compensate uncertainty and process variations. Most performance improvement can be done simply by keeping certain constraints active, in this case the terminal constraints, using run-to-run adaptation. Also, the improvement in cost was obtained in 2-4 batches, showing that the success of the method depends on a well-designed adaptation law.

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