Proceedings of the 17th World Congress
The International Federation of Automatic Control
Seoul, Korea, July 6-11, 2008

On-line Optimizing Control for a Class of Batch Reactive Distillation Columns

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Abstract: The problem of designing an on-line optimizing output-feedback (OF) controller for the class of reaction: A + B ↔ C + D of batch reactive distillation columns with temperature measurements is addressed. The joint process-control design problem is solved within a constructive framework, by combining relative degree and detectability structures concepts in the light of particular system features. The result is an OF control scheme that decides the total reflux period and batch durations, and manipulates the reflux rate over the withdrawal period. The proposed approach is illustrated with a case example (esterification of ethanol and acetic acid) through numerical simulations, yielding a closed-loop operation which is similar to the open-loop ones previously obtained via direct optimization.

1. INTRODUCTION

Reactive distillation (RD) is a process where reaction and separation are carried out in the same unit, provided that the product of interest has the largest or lowest boiling point (Taylor and Krishna, 2000). The advantages of RD are reduced investment and operating costs, environmental impacts, and so on, due to: (i) increased yield of a reversible reaction by separating the product of interest from the reaction mixture, (ii) overcome, by chemical reaction, of thermodynamic separation limitations (Sundmacher and Kienle, 2002). Most of the published works in RD have focused to continuous columns, and less attention has been given to the batch mode. While the open-loop operation design of batch RD columns has been successfully addressed via optimization techniques (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001), the study of the associated tracking controller lags behind.

On the other hand, temperature sensor location criteria from conventional (i.e. non-reactive) distillation columns have been applied to the RD case (Venkateswarlu and Kumar, 2006), and model-based state estimation studies have been performed with the extended Kalman filter (EKF) (Wilson and Martinez, 1997). The OF controllers have been designed with linear (PI, PID) (Sørensen and Skogestad, 1994; Monroy-Loperena and Alvarez-Ramirez, 2000; Georgiadis *et al.*, 2002) and nonlinear MPC (*Model Predictive Control*; Balasubramhanya and Doyle III, 2000; Engell and Ferhonlz, 2003) techniques to track temperature or product composition by manipulating reflux rate or heat duty. The control part of the problem has been separately addressed from the optimal operation design part. The joint operation-control design problem of an experimental semibatch column system has been addressed with nonlinear dynamic optimization techniques (Noeres *et al.*, 2004) with emphasis on the optimal open-loop operation and control structure aspects. This study demonstrates the feasibility of jointly addressing the process-control design problem. However, the design procedure is still rather complex and disconnected from the control design, and it is not clear to what extent the control structure results depend on the particular control scheme employed and its tuning.

Recently, a joint operation-control constructive design approach for batch processes has been proposed and applied to emulsion polymerization (Alvarez *et al.*, 2004) and binary distillation columns (Alvarez *et al.*, 2005), exploiting specific system structural properties such as relative degrees and detectability. Instead of performing the operation optimization in open-loop mode, the same task is performed over the process passive dynamical inverse, or equivalently, the limiting closed-loop behavior attainable with robust state-feedback (SF) control. Then, the output-feedback (OF) tracking controller is constructed by redesigning the feedback controller associated to the optimal dynamical inverse and incorporating a state-estimator. By doing so, the optimal motion and robust control designs are closely connected. These considerations motivate the present study.

In this work, the problem of designing an on-line optimizing OF controller for the class A + B ↔ C + D of batch RD columns with temperature measurements is addressed within a constructive framework (Sepulchre *et al.*, 1997). The result is an OF control scheme that decides the total reflux period, manipulates the reflux over the withdrawal period, and stops the operation when a profit index is maximized. The proposed approach is illustrated with a case example (esterification of ethanol and acetic acid) through numerical simulations. The resulting closed-loop batch operation resembles to the open-loop ones drawn before (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001) with standard optimization techniques for the process design part of the problem.

2. CONTROL PROBLEM

Consider the N-tray batch RD column depicted in Figure 1, equipped with a reboiler, a total condenser, an accumulator vessel and temperature measurements (T_j) in the s_j-th stages (from reboiler to N-tray); a catalyzed reversible reaction takes place ($\alpha_A A + \alpha_B B \overset{H+}{\leftrightarrows} \alpha_C C + \alpha_D D$), and the product of interest (C) has the lowest boiling point. The thermodynamic and kinetic functions are given by

$$\nu(c) = (\nu_A, \nu_B, \nu_C)'(c), \qquad T = \beta(c) \qquad c = (c_A, c_B, c_C)' \qquad (1a)$$
$$r(c, T) = \rho_M[k_1(T)c_A c_B - k_2(T)c_C c_D], \quad c_D = 1 - c_A - c_B - c_C, \quad (1b)$$
$$\rho(c) := r[c, \beta(c)], \qquad \rho_l(c) = \alpha_l \rho(c), \quad l = A, B, C \quad (1c)$$
$$\nu_D(c) = 1 - \nu_A(c) - \nu_B(c) - \nu_C(c) \qquad (1d)$$

where c_l is the l-th mole fraction in the liquid, $\nu(c)$ is the liquid-vapor equilibrium function that determines the vapor compositions, β is the bubble point function that sets the mixture temperature T, ρ_M is the mixture molar density, ρ is the total reaction rate per unit of time, α_l is the l-th stoichiometric coefficient, and ρ_l is the l-th component reaction rate.

The column is operated as follows (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001): at time t = 0, a mixture of m_L moles at compositions c_{Ao}, c_{Bo}, and c_{Co} of reactants A, B, and product C, is loaded, catalyst (H^+) is added, and heat is supplied at a constant rate $Q \in [Q^-, Q^+]$ set by equipment design and operation considerations. During the start-up period $[0, t_r]$, the column is operated at total reflux (molar) rate ($R = V_N$) until a time t_r when the product C reaches a certain purity. Then, in the withdrawal period $(t_r, t_f]$, the reflux rate R is adjusted to maintain a constant (at certain optimal value) product purity in the time-varying distillate flowrate $\mathcal{D} = V_N - R$, until the final time t_f.

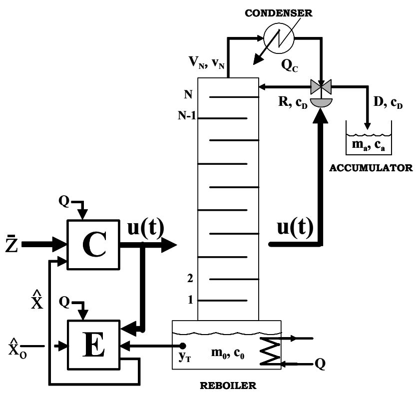

Figure 1. Batch RD column and control system.
C: Controller, **E**: State Estimator

The heat duty (Q), the accumulator product purity (c_a^C), the reflux (or final) time t_r (or t_f), and the reflux rate function R(t) over $(t_r, t_f]$ must be chosen to maximize the batch time-average profit (\$/time):

$$J = \{k_p c_a^C m_a(t_f) - k_L - k_h Q t_f - \int_0^{t_f} K_c Q_c(\tau)]d\tau\}/(t_f + t_d) - k_O \qquad (2)$$
$$k_L = (k_A m_{Ao} + k_B m_{Bo} + k_p m_{Co} + k_D m_{Do})$$

where $m_a(t_f)$ is the final accumulator molar mass, k_p is the product value per mole, m_{Ao} (or m_{Bo}, m_{Co}, m_{Do}) is the loaded molar mass of component A (or B, C, D) and k_A (or k_B, k_D) is its cost per mole, k_h (or K_c) is the heating (or cooling) cost per heat unit, Q_c is the time-varying heat removal rate in the condenser, k_O is the operation cost per time unit, and t_d is the dead time between batches. Under standard assumptions (reaction only in the liquid phase, vapor-liquid equilibrium at each stage, negligible vapor holdup, pressure drop, mixing heat and energy loss, and quasi-steady state regime for enthalpy and molar tray holdup dynamics; Cuille and Reklaitis, 1986; Mujtaba and Macchietto, 1997), the column behavior is described by the following dynamic equations over the batch operation period $[0, t_f]$:

• *Compositions* $[c_k = (c_k^A, c_k^B, c_k^C)', \; k = 0, ..., N, \mathcal{D}, a]$

$$\dot{c}_0 = \{L_1(c_1 - c_0) - V_0[\nu(c_0) - c_0]\}/m_0 + \rho_T(c_0), \qquad (3a)$$
$$\dot{c}_i = \{L_{i+1}(c_{i+1} - c_i) + V_{i-1}[\nu(c_{i-1}) - c_i] - V_i[\nu(c_i) - c_i]\}/m_i + \rho_T(c_i), \quad 1 \le i \le N-1, \quad \rho_T = (\rho_A, \rho_B, \rho_C)' \qquad (3b)$$
$$\dot{c}_N = \{(R - V_N)[\nu(c_N) - c_N] + V_{N-1}[\nu(c_{N-1}) - c_N)]\}/m_N + \rho_T(c_N), \qquad c_{\mathcal{D}} = \nu(c_N) \qquad (3c)$$
$$\dot{c}_a = 0 \quad \forall\, t \le t_r, \quad \{\text{ or } = (V_N - R)[\nu(c_N) - c_a]/m_a \quad \forall\, t > t_r\} \qquad (3d)$$
$$y_j = T_j = \beta(c_j), \qquad c_j(0) = c_{jo}, \quad 0 \le j \le N \qquad c_a(t_r) = 0$$

• *Liquid Holdups*

$$\dot{m}_0 = L_1 - V_0, \qquad m_0(0) = m_{0o} \qquad (3e)$$
$$\dot{m}_a = 0 \quad \forall\, t \le t_r, \quad \{\text{ or } = V_N - R \quad \forall\, t > t_r, \; m_a(0) = m_{ao}\} \qquad (3f)$$

• *Profit*

$$\dot{\varphi} = [k_p \nu_C(c_N)(V_N - R) - (\varphi + k_h Q + K_c Q_c + k_O)]/(t + t_d) := f_\varphi \qquad (3g)$$
$$J = \varphi(t_f), \qquad \varphi_0 = -(k_L/t_d + k_O)$$

• *Energy balances and tray hydraulics*

$$V_0 = [L_1(h_1 - h_0) + Q + \theta_R(c_0, m_0)]/\lambda_0 \qquad (3h)$$
$$V_i = [L_{i+1}(h_{i+1} - h_i) + V_{i-1}(H_{i-1} - h_i) + \theta_R(c_i, m_i)]/\lambda_i \qquad (3i)$$
$$V_N = [R(h_{\mathcal{D}} - h_N) + V_{N-1}(H_{N-1} - h_N) + \theta_R(c_N, m_N)]/\lambda_N := \upsilon_N \qquad (3j)$$
$$L_i = R - V_N + V_{i-1}, \quad L_i = a_\eta(m_i - b_\eta)^{c_\eta} := \eta(m_i), \quad 1 \le i \le N, \qquad (3k)$$
$$\theta_R(c_j, m_j) := m_j[\delta_r(c_j)]\rho(c_j),$$
$$\Delta H_r(T_j) = \Delta H_r[\beta(c_j)] := \delta_r(c_j), \quad 0 \le j \le N$$
$$R = V_N, \quad 0 \le t \le t_r; \qquad \text{or} \qquad R \le V_N, \quad t_r < t \le t_f$$
$$Q \in [Q^-, Q^+], \qquad Q_c = V_N \lambda_c$$

where c_k is the vector of compositions in the liquid at the k-th stage, m_k is the liquid holdup at the k-th stage, η is the Francis' hydraulics function, λ_k (or λ_c) is the vaporization latent heat (or condensation) at k-th stage (or condenser), h_k (or H_k) is the enthalpy of the liquid (or vapor) at k-th stage, θ_R is the reaction heat function, and V_k (or L_k) is the vapor flow rate (or liquid flow rate) at k-th stage. In compact vector notation the *column model* is given by the nonautonomous system:

$$\dot{x} = f(x, d, u), \qquad x(0) = x_0, \qquad y = h(x), \qquad t \in [0, t_f] \quad (4)$$
$$x = (c_0', ..., c_N', c_a', m_0, m_a, \varphi)', \qquad \dim(x) = n = 3(N+2) + 3$$
$$f = (f_{c_0}', ..., f_{c_N}', f_{c_a}', f_{m_0}, f_{m_a}, f_\varphi), \qquad u = R, \qquad d = Q$$

$$h(x) = [\beta(c^A_{S_1}, c^B_{S_1}, c^C_{S_1}), \ldots, \beta(c^A_{S_m}, c^B_{S_m}, c^C_{S_m})]$$

where x is the state, u (or d) control (or exogenous) input, and y is the measured output. The solution state motion x(t) and the corresponding output trajectory y(t) are denoted by

$$x(t) = \tau_x[t, x_o, d, u(\cdot)], \tag{5a}$$

$$y(t) = h[x(t)] = h\{\tau_x[t, x_o, d, u(\cdot)]\} = \tau_y[t, x_o, d, u(\cdot)] \tag{5b}$$

The standard definitions of steady-state asymptotic (infinite time) stability for continuous processes, do not apply to the (finite time) batch motion case, and the same is true for controllability and detectability. In industrial practice, a batch operation is regarded "stable" if admissible initial and exogenous input disturbance sizes produce admissible state motion and output trajectory deviation sizes (Alvarez *et al.*, 2004a). These stability notions correspond to the ones of practical input-to-state and input-to-output stability (La Salle and Lefschetz, 1961; Sontag, 2000). For the batch case, the stability is stated next. The *motion* x(t) (5a) is *practically (P) stable* if, for given disturbance-response sizes (δ_o, δ_d, δ_u, ε_x), there are positive constants (a_x, λ_x, γ^x_d, γ^x_u) so that:

$$|\tilde{x}_o| \le \delta_o, \quad ||\tilde{d}(t)|| \le \delta_d, \quad ||\tilde{u}(t)|| \le \delta_u \Rightarrow |\tilde{x}(t)| \le a_x e^{\lambda_x t}|\tilde{x}_o| \tag{6a}$$
$$+ \gamma^x_d||\tilde{d}(t)|| + \gamma^x_u||\tilde{u}(t)|| \le a_x\delta_o + \gamma^x_d\delta_d + \gamma^x_u\delta_u = \varepsilon_x, \quad ||(\cdot)(t)|| = \sup_t |(\cdot)|$$

where $|(\cdot)|$ is the Euclidean norm of the vector $(\cdot)$. The *output trajectory* y(t) (5b) is *P-stable* if, for given (δ_o, δ_d, δ_u, ε_y), there are (a_y, λ_y, γ^y_d, γ^y_u) so that

$$|\tilde{x}_o| \le \delta_o, \quad ||\tilde{d}(t)|| \le \delta_d, \quad ||\tilde{u}(t)|| \le \delta_u \Rightarrow |\tilde{y}(t)| \le a_y e^{\lambda_y t}|\tilde{x}_o| \tag{6b}$$
$$+ \gamma^y_d||\tilde{d}(t)|| + \gamma^y_u||\tilde{u}(t)|| \le a_y\delta_o + \gamma^y_d\delta_d + \gamma^y_u\delta_u = \varepsilon_y$$

In our batch RD column we are interested in an optimal closed-loop operation with P-stable motion, and above all, "output profit" $y_\varphi(t) = \varphi(t)$ P-stable output, with emphasis on its final time value $y_\varphi(t_f) = J$.

Given the model (4), our *problem* consists in designing:

(i) The nominal optimal operation

$$\mathbf{O}: \{\bar{d}, \bar{u}(t), \bar{x}(t), \bar{t}_r, \bar{t}_f\} \tag{7}$$

so that the batch ends with *maximum profit* $J = \varphi(t_f)$, and the operation **O** is closed-loop (state and output) P-stable with OF control (8).

(ii) The robust dynamic OF tracking controller

$$\mathbf{C}: \dot{x}_c = f_c(x_c, d, y), \quad x_c(0) = x_{co} \approx \bar{x}_{co}, \quad t \in [0, t_f] \tag{8a}$$
$$t_r = \mu_r(x_c, d) \quad u = \mu(x_c, d), \quad t_f = \mu_f(x_c, d) \tag{8b-d}$$

that, driven by temperature measurements (in stages to be determined), decides: the total reflux period (t_r), the reflux rate policy over the withdrawal period (t_r, t_f], and the batch termination time (t_f).

3. NOMINAL CLOSED-LOOP OPERATION

In previous studies (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001; Noeres *et al.*, 2004), the optimal operation **O** (7) has been drawn by finding the set [Q, R(t), t_r, t_f] that maximizes the profit J (2) subjected to the *open-loop column dynamics* (4), but it is unclear the connection with the feedback control design. Following the approach employed before in polymerization reactors (Alvarez *et al.*, 2004a) and binary distillation columns (Alvarez *et al.*, 2005), here the optimal motion problem will be addressed in terms of the closed-loop system with passive (robustness-oriented) SF control, with the understanding that optimal SF controllers are: (i) nonwasteful, (ii) inherently robust, and (iii) passive (relative degree less or equal to 1 and minimum phase) with respect to a certain output z (Sepulchre *et al.*, 1997).

In the *total reflux regime* [R = V_N = $\upsilon_N(x, d)$], the corresponding column dynamics are given by (9), and its solution motion is denoted by (10):

$$\dot{x} = f[x, d, \upsilon_N(x, d)], \quad x(0) = x_0, \quad y = h(x), \quad t \in [0, t_r] \tag{9}$$

$$x(t) = \tau_r[t, x_0, d] \tag{10}$$

3.1 Reflux control over the withdrawal period

The profit J (2) dependency on the accumulator product composition (c^C_a), and results from previous RD column studies via open-loop optimization (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001) suggest the consideration of the N-th tray vapor composition $\nu_c(c_N)$ as regulated output at a fixed value $\bar{z}$ (to be determined) smaller than the value z* asymptotically reached in total reflux regime. This is (rd := relative degree),

$$z = \nu_c(c_N), \qquad rd(z, u) = 1, \qquad z \in [0, z^*] \tag{11}$$

provided: (i) the rd = 1 conditions (12a-b) are met, or equivalently, the algebraic equation pair (13a-b) has a unique solution (14a-b) for (c^C_N, u), and (ii) the corresponding inverse (or zero-dynamics) motion is P-stable,

$$\forall\, t \in (t_r, t_f]: \quad \partial\nu_C(c_N)/\partial c^C_N > 0, \quad \partial\gamma(x_I, d, u)/\partial u > 0 \tag{12a-b}$$

$$\nu_c(c_N) = z, \qquad \gamma(x_I, d, u) = 0 \tag{13a-b}$$

$$c^C_N = \sigma(z, c^A_N, c^B_N), \qquad u = \mu_I(x_I, d, z) \tag{14a-b}$$

where the maps γ, σ, and μ_I are given by

$$\gamma(x_I, d, u) = [\partial_{C^A_N}\nu_C(c_N)]f_{C^A_N} + [\partial_{C^B_N}\nu_C(c_N)]f_{C^B_N} + [\partial_{C^C_N}\nu_C(c_N)]f_{C^C_N}$$
$$\sigma(z, c^A_N, c^B_N) = \nu^{-1}_C(z, c^A_N, c^B_N)$$

$$\mu_I(x_I, d, z) = \gamma^{-1}(x_I, d, z, \dot{z} = 0)$$
$$x_I = (c'_0, \ldots, c'_{N-1}, c^A_N, c^B_N, c'_a, m_0, m_a, \varphi)', \qquad \dim x_I = n-1$$

γ^{-1} is the inverse of γ for u, and x_I is the state of the dynamical inverse (Hirschorn, 1979):

$$\dot{x}_I = f_I(x_I, d, u), \qquad x_I(t_r) = x_{Ir} \qquad t \in (t_r, t_f] \tag{15a}$$
$$u = \mu_I(x_I, d, z), \qquad x_z = \sigma(x_I, z) \tag{15b-c}$$

where (I_x is an identity permuted matrix)

$$I_x[x'_{Ir}, x_z(t_r)]' = \tau_x[t_r, \bar{x}_0, d, u(\cdot)], \qquad x_z = c^C_N$$
$$f_I = (f_{c'_0}, \ldots, f_{c'_{N-1}}, f_{C^A_N}, f_{C^B_N}, f_{c'_a}, f_{m_0}, f_{m_a}, f_\varphi)', \qquad c^C_N = \sigma(z, c^A_N, c^B_N)$$
$$\tau_x[t_r, \bar{x}_0, d, u(\cdot)] = I_x[x'_{Ir}, x_z(t_r)]', \qquad x_z(t_r) = \sigma[x_I(t_r), z]$$

The solution of (15a) is the dynamical inverse motion (16), and the inverse state motion is given by (17a-b):

$$x_I(t) = \tau_x[t, x_{Ir}, d, u(\cdot)], \qquad t \in (t_r, t_f] \qquad (16)$$
$$x(t) = I_x[x_I'(t), x_z(t)]', \qquad x_z(t) = \sigma[x_I(t), z] \qquad (17a\text{-}b)$$

Summarizing, given final state $[x_I(t_r)]$ of the total reflux period, the heat duty (Q), and the distillate composition (z), the integration of the dynamical inverse (15a) yields the reflux rate over $t \in (t_r, t_f]$ in the feedback control form (15b).

3.2 Total reflux period and batch durations

The total reflux period ($R = V_N$) ends at time t_r, when the vapor composition $\nu_c(c_N)$ reaches the prescribed value z , and the batch finishes at time t_f, when the time-average profit function $\varphi(t)$ (3g) reaches its maximum value J (2). This yields the *events SF controller*:

$$t_r = \{t \in \Re \mid \nu_c[c_N(t)] = z\} := \mu_r(x, z) \qquad (18a)$$
$$t_f = \{t \in \Re \mid f_\varphi[x(t), d, u(t), t] = 0\} := \mu_f(x, d, u) \qquad (18b)$$

3.3 Maximum profit J with respect to (d, z)

Hitherto, for a given (heat duty, N-th tray vapor composition) pair (d, z), we have obtained the nominal operation (17) in closed-loop form with passive nonlinear SF control (15b).

Thus, the optimal pair $(\bar{d}, \bar{z})$ can be obtained by solving the straightforward (algebraic) maximization problem

$$(d, z) = (\bar{d}, \bar{z}) \ni \max_{(d,z)} J, \quad d \in [Q^-, Q^+], \quad z \in [0, z^*] \qquad (19)$$

3.4 Optimal closed-loop operation

The combination of the total reflux (10) and withdrawal (16) motions, with their pair (d, z) set at its optimal value (19), yields the optimal closed-loop operation **O** (20):

- *Total reflux period $[0, t_r]$:*

$$\bar{x}(t) = \tau_r(t, t_o, \bar{x}_o, \bar{d}), \quad \bar{u}(t) = \upsilon_N[\bar{x}(t), \bar{d}], \quad \bar{t}_r := \mu_r(\bar{x}, \bar{z}) \qquad (20a\text{-}c)$$

- *Withdrawal period $(t_r, t_f]$:*

$$\bar{x}(t) = I_x[\bar{x}_I'(t), \bar{x}_z(t)]', \quad \bar{x}_I(t) = \tau_x(t, t_o, \bar{x}_o, \bar{d}), \quad \bar{x}_z(t) = \sigma[\bar{x}_I(t), \bar{z}],$$

$$\bar{u}(t) = \mu_I(\bar{x}_I, \bar{d}, \bar{z}), \qquad \bar{t}_f = \mu_f[\bar{x}(t), \bar{d}, \bar{u}(t)] \qquad (20d\text{-}h)$$

where the motion x(t) and the output profit $\varphi(t)$ must meet certain P-stability requirements, which can be characterized by straightforward simulation.

4. OUTPUT-FEEDBACK OPTIMIZING CONTROL

In this section, the OF tracking controller is constructed as follows: (i) first, the SF controller (14a-b) of the optimal dynamical passive inverse (15a) is redesigned to compensate initial state and exogenous input deviations, (ii) then, a robust passive state estimator is designed as an extension of the one presented for a ternary distillation columns (Pulis, 2007), and (iii) the SF controller and estimators are combined.

4.1 SF optimizing controller

From the enforcement of the closed-loop output tracking error linear dynamics (22a) upon the open-loop column dynamics (4) the passive *SF tracking controller* (22b) follows (k_c is the adjustable control gain):

$$\dot{\tilde{z}} = - k_c\tilde{z}, \qquad \tilde{z} = z - \bar{z}, \qquad t \in (t_r, t_f] \Rightarrow \qquad (22a)$$

$$u = \gamma^{-1}\{x_I, \bar{d}, \nu_c(c_N), -k_c[\nu_c(c_N) - \bar{z}]\} := \mu(x, \bar{d}, \bar{z}) \qquad (22b)$$

which is a redesigned version, to compensate for initial state and exogenous input deviations, of the SF controller (15b) which underlies the optimal closed-loop operation (20). The combination of this controller with the event component controllers (18a-b) yields the *on-line optimizing SF controller*

$$t \in [0, t_r]: \quad u(t) = R(t) = \upsilon_N(x, \bar{d}), \qquad t_r = \mu_r(x, \bar{z}) \qquad (23a)$$

$$t \in (t_r, t_f]: \quad u(t) = R(t) = \mu(x, \bar{d}, \bar{z}), \qquad t_f = \mu_f(x, \bar{d}, \bar{z}) \qquad (23b)$$

whose behavior: (i) represents the one attainable with any observer-based passive OF controller, and (ii) will be considered the recovery target for our OF design.

4.2 OF dynamic controller

Following the constructive estimation technique (Lopez and Alvarez, 2004; Fernandez and Alvarez, 2007) and its application to binary (Alvarez *et al.*, 2005) and ternary (Pulis, 2007) distillation columns, let us regard a robustness-oriented m-temperature passive estimator for our RD column (with relative degree *1* between the measured output and its integral state, *meaning one innovated state per measurement*):

$$\dot{\hat{x}}_\iota = f_\iota[\hat{x}, u(t)] + O^{-1}(\hat{x})\{\hat{\iota} + K_y[y(t) - h(\hat{x})]\}, \quad \hat{x}_\iota(0) = \hat{x}_{\iota o} \qquad (24a)$$
$$\hat{x}_\iota = (\hat{c}^l_{s_1}, ..., \hat{c}^l_{s_m})', \quad l = A, B, C, \quad \dim \hat{x}_\iota = \dim \hat{\iota} = m$$

$$\dot{\hat{\iota}} = K_\iota[y(t) - h(\hat{x})], \qquad \hat{\iota}(t_0) = 0 \qquad (24b)$$

$$\dot{\hat{x}}_v = f_v[\hat{x}, u(t)], \qquad \hat{x}_v(0) = \hat{x}_{vo}, \qquad \dim \hat{x}_v = n - m \qquad (24c)$$

where

$$(\hat{x}_\iota', \hat{x}_v')' = I_p\hat{x}, \qquad (f_\iota', f_v')' = I_p f$$
$$O(\hat{x}) = \mathrm{diag}[\beta_{c_l}(\hat{c}^A_{s_1},\hat{c}^B_{s_1},\hat{c}^C_{s_1}), ..., \beta_{c_l}(\hat{c}^A_{s_m},\hat{c}^B_{s_m},\hat{c}^C_{s_m})]'$$
$$\beta_{c_l}(\hat{c}_A, \hat{c}_B, \hat{c}_C) = \partial\beta/\partial c_l$$
$$K_y = \mathrm{diag}[2\zeta_1\omega_1, ..., 2\zeta_m\omega_m], \qquad K_\iota = \mathrm{diag}[\omega_1^2, ..., \omega_m^2]$$

x_ι (or x_v) is the innovated (or non-innovated) state, ι is an integral state to eliminate output mismatch, O is the estimation matrix, I_p is a column-permuted identity matrix, and ω_i (or ζ_i) is the characteristic frequency (or damping factor) of the i-th output estimation error dynamics. The determination of the number (m) of sensors and locations of the temperature sensors, and the innovated state choice can be performed with sensitivity measures, temperature gradients and their partition into component contributions, according to the straightforward application of procedures employed in previous ternary distillation column estimation (Pulis, 2007) and binary distillation column optimal operation-control (Alvarez *et al.*, 2005) studies.

The combination of the preceding geometric estimator (24) with the optimizing SF controller (23) yields a dynamic OF controller of the form (8), with the tuning guidelines given in (Alvarez *et al.*, 2005). The rigorous verification of the related closed-loop stability conditions goes beyond the scope of the present work, and here it suffices to state that P-stability of the closed-loop motion requires sufficient dynamic separation between the observer (ω_i) and control (k_c) gains, and that the observer gain is limited by the high frequency column unmodeled (hydraulics and enthalpy) dynamics in conjunction with the measurement instrument error.

4.3 Concluding remarks

By construction, the optimal motion and its tracking OF control designs are connected via the dynamical inverse concept which: (i) is part of the dynamic restriction in the nominal optimal closed-loop design algorithm, (ii) sets the limiting closed-loop behavior attainable with robust SF control, and (ii) is the point of departure for the construction of the robust OF tracking controller.

5. CASE STUDY

To test the proposed operation-control design approach, the esterification reaction: ethanol (A) + acetic acid (B) $\overset{H+}{\leftrightarrows}$ ethyl acetate (C) + water (D), catalyzed with sulphuric acid, was chosen as application example via simulations, with the understanding that the optimal operation part of our problem has been studied before with open-loop optimization (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001). The kinetics and the thermodynamic model and parameters were taken from Georgiadis *et al.* (2002) and Giessler *et al.* (1999), respectively. The column model describes an experimental glass batch column (Fernández and Alvarez, 2007) with N = 10 trays, minimum-maximum heat duty (Q^-, Q^+) = (0.6, 2) kW, hydraulic parameter set (a_η, b_η, c_η) = (3.0, 0.2, 1.5), 1.5 L reboiler volume, and 0.02 L tray volume. The column was loaded with 30.25 moles at composition $c_{j0} = (c_{Ao}, c_{Bo}, c_{Co})' = (0.45, 0.51, 0)'$. The objective function (2) constants are: (k_p, k_L) = (4, 0.02245)\$/mol, (k_h, K_c) = (2.8×10^{-9}, 4.7×10^{-10})\$/J, k_o = 0.01241 \$/h, and t_d = 0.5 h.

5.1 Closed-loop operation with SF optimizing control

The application of the design procedure presented in Section 3 yielded ($\bar{Q}$, $\bar{z}$) = (1.2 kW, 0.5646), ($\bar{t}_r$, $\bar{t}_f$) = (1.7, 5.75)h. Basically, the N-th tray vapor composition profile is not appreciably modified by changes in the heat duty (Q), but the optimal value $\bar{Q}$ yields more profit (about 16 %) than the one obtained with Q^+. The optimal composition value ($\bar{z}$) for the product of interest was chosen as high as possible and safely away from reflux control saturation. The control gain was set $k_c = \lambda_m/8 = 1/40$ min^{-1}, where $\lambda_m = 1/(0.5\ N)$ min^{-1} is the individual tray characteristic frequency of the hydraulic-enthalpy dynamics. To preclude initial wasteful R(t) control action (in the switching from total-reflux period to withdrawal period), at a minor cost in time, switching of the nominal optimal operation was smoothed by SF control shown in Figure 2 (continuous plots). In the same figure are included results for three perturbed load composition sets (discontinuous plots). The control scheme is able to modify the decisions on t_r, t_f and R, to maximize the profit J, and in all cases the optimal product value $\bar{z}$ is well tracked. It must be pointed out that, basically, the nominal closed-loop optimal motion (Figure 2) with analytic SF passive nonlinear controller resembles the ones drawn before with open-loop optimization (Mujtaba and Macchietto, 1997; Giessler *et al.*, 2001).

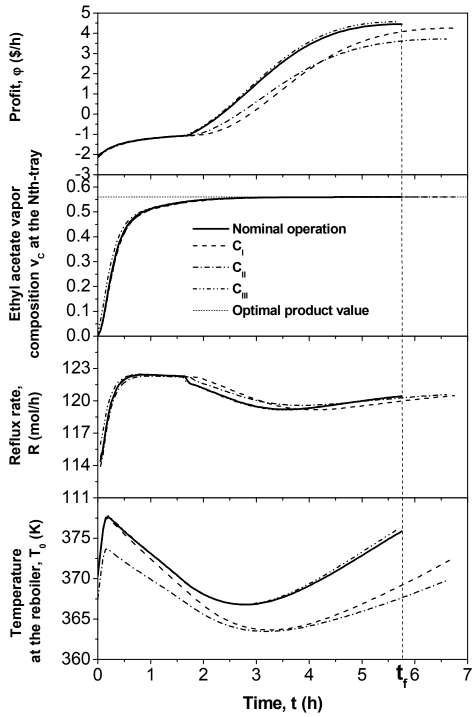

Figure 2. Closed-loop operation with optimizing SF control, for various load compositions sets: nominal $(c_{Ao}, c_{Bo}, c_{Co})' = (0.45, 0.51, 0)'$, and perturbed $C_I = (0.5, 0.5, 0)'$, $C_{II} = (0.45, 0.45, 0)'$, and $C_{III} = (0.45, 0.51, 0.006)'$.

That it is not possible to obtain pure product in this esterification case, because azeotrope boiling points are slightly lower than the desired product (ethyl acetate) is a known fact from previous studies (Giessler *et al.* 1999).

5.2 Closed-loop operation with OF optimizing control

With respect to the OF control implementation, the application of the sensor selection and innovated state choice criteria previously presented (Pulis, 2007) yielded: (i) a reasonable behavior with a single measurement in the reboiler, and (ii) the acetic acid composition in the reboiler (c_0^B) as the innovated state (x_ι). The application of the tuning guidelines discussed in Section 4 yielded: $\zeta_i = 2.0$, $\omega_i = \lambda_m/3 = 1/15$ min^{-1} (and the previous k_c= 1/40 min^{-1}), meaning that the observer was tuned about three times faster than the controller, or three times slower than the hydraulic-enthalpy parasitic dynamics. The estimator was run with -5 % initial estimate errors. The related results in Figure 3 show that: (i) as expected, the OF controller undergoes some degradation in comparison with its SF counterpart, and (ii) basically, the OF controller recovers reasonably well the behavior of the SF controller, for the nominal and perturbed loads.

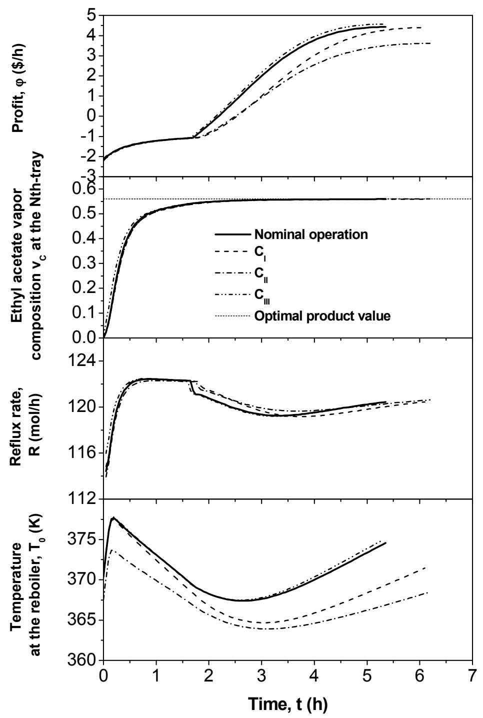

Figure 3. Closed-loop operation with optimizing OF control, for various load compositions sets: nominal and perturbed C_I, C_{II}, C_{III} (listed in Figure 2).

6. CONCLUSIONS

The problem of designing an on-line optimizing output-feedback (OF) controller for the class $A + B \leftrightarrow C + D$ of batch RD columns with temperature measurements has been addressed. The combined optimal operation-control problem was solved with an interlaced robustness-oriented estimator-control scheme, by introducing a profit state and exploiting the relative degree and detectability system properties. This enabled the connection between the optimal operation and OF control designs, and their treatment within a unified framework. The proposed approach is illustrated with a case example (esterification of ethanol and acetic acid) through numerical simulations, finding that: (i) the closed-loop optimal operation with robust OF control resembled the ones drawn before with open-loop optimization, and (ii) the on-line optimizing OF tracking controller was rather robust and effectively manipulated the reflux rate to track the nominal motion, with only a temperature measurement in the reboiler.

Acknowledgments. The authors gratefully acknowledge the support from CONACyT: PhD Scholarship (157353) for S. Pérez-Correa, and research grant (Design and Control of Batch Processes, 47927).

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