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Control of a reactive batch distillation process using an iterative learning technique

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Abstract−Quadratic criterion-based iterative learning control (QILC) was applied to a numerical reactive batch distillation process, in which methacrylic anhydride (MAN) is produced through the reaction of methacrylic acid with acetic anhydride. The role of distillation is to shift the equilibrium conversion toward the direction of the product by removing acetic acid (AcH), a by-product of the reaction. Two temperatures at both ends of the column were controlled by individual control loops. A nonlinear PID controller manipulating the reflux ratio was employed to regulate the top temperature at the boiling point of AcH. A constrained QILC was used for the tracking of the reactor temperature. A time-varying reference trajectory for the reactor temperature that satisfies the target conversion and purity of MAN was obtained through repeated simulations and confrimation experiments in the pilot plant. The QILC achieved satisfactory tracking in several batch runs with gentle control movements, while the PID control as a substitute of the QILC in a comparative study exhibited unacceptable performance.

Keywords: Iterative Learning Control, QILC, Reactive Batch Distillation, Batch Distillation Control

INTRODUCTION

Batch distillation (BD) is a process that runs under an unsteady state over an entire batch horizon and poses challenging but interesting operational issues. The most important challenge is to find optimum trajectories for process variables that minimize the operating cost while satisfying various product specifications. Achieving the trajectories by manipulating the boil-up and reflux rates, based on the measurement feedback, is another challenge. Both problems are non-trivial and have been the focus of many research studies. The present study is devoted to designing a control system for a reactive batch distillation (RBD) process.

A batch distillation unit is often designed as a part of a batch reactor for product or by-product removal to shift the equilibrium conversion. Such a distillation-reactor combination is a typical configuration of RBD processes. The RBD process shares the same operational issues as the BD process, except that the reactor temperature is determined in consideration of the reaction as well as the separation. The temperature should be constrained for thermal degradation of the products and/or undesired side reactions not to occur. The heat input to the reactor needs to be constrained for the column flooding and/or local hot spots on the reactor wall to be properly inhibited. All these considerations together with the regulation and tracking requirements render the control of an RBD process a challenging task.

Despite its importance, a literature survey revealed that quite limited studies have been published so far on the optimization and control of BD and/or RBD processes. One conceivable reason is the difficulty in developing a realistic numerical model of a BD or RBD process. Most of the papers that are cited in this study considered simplified process models comprised of mass balance equations under the equi-molar overflow assumption, idealized vapor-liquid equilibrium relationship, and simple kinetic equations. Recently, the environment for numerical study has changed greatly as commercial simulators for batch distillation processes, which are dynamic in nature, have become available.

Sorensen et al. [1] conducted an experimental study of the optimal operation of an RBD process that produces poly-terephthalate through condensation polymerization of a dibasic aromatic acid with two glycols. The role of distillation is to remove water to increase the product yield. The configuration and purpose of the process were similar to those considered in this study. An optimal constant reboiler heat input and time-varying reflux rate were computed using a simplified process model. To implement the optimal operating strategy, the temperature of the third tray from the top was controlled using a PI controller with the optimal temperature trajectory as the set point and the reflux rate as the manipulated variable (MV).

Mujtaba et al. [2] discussed the optimization of an RBD process using a polynomial curve-fitting technique. The process under investigation produced ethyl acetate from acetic acid and ethanol. Ethyl acetate was removed from the top of the column so that the reaction could continue to completion. The time-varying reflux rate and reboiler heat input trajectories were represented as polynomials in time, and their coefficients were determined through optimization. The RBD process was described using simplified model equations.

Fileti et al. [3] applied different types of controllers to a BD process to overcome the nonlinearity that arises during the batch operation. They considered gain-scheduled PI control, a self-tuning regulator, and a non-linear model predictive control (MPC), and evaluated the performance of each control method in a pilot BD column separating a mixture of n-hexane, benzene and toluene.

Balasubramhanya and Doyle III [4] considered the same RBD pro-

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cess as in Mujtaba et al. [2], and proposed a nonlinear MPC (NMPC) method using a reduced model. The process model was reduced to a set of five differential equations and six algebraic equations. The NMPC method was designed to regulate the top temperature at a set point that maximizes the process productivity.

Model-based control methods, such as MPC [5], can provide good tracking performance with modest control movements if a reliable process model is available. Identifying such a model for a BD or RBD process is not generally easy because the process is operated under unsteady state conditions, and severe nonlinearity may arise during the batch run.

Iterative learning control (ILC) is a batch process control technqiue that can overcome the drawback of model-based real-time control methods. Using the batch-wise integral action, ILC can achieve perfect tracking as the batch run repeats even when there is a significant uncertainty in the model [6]. The BD and RBD operations are batchwise, and might be greatly improved using the ILC.

We propose a control system for an RBD process and investigate its performance. The process under investigation produces methacrylic anhydride (MAN) in the reactor vessel, while continuously removing acetic acid (AcH) from the top of the distillation column. The RBD process was numerically realized using the Batch Distillation module in ASPEN Plus^{TM}, whose performance was confirmed through experiments in a pilot plant of Samsung Cheil Industries, Inc. The top temperature of the column was regulated at the boiling point of AcH by using a nonlinear PID controller manipulating the reflux ratio. A constrained QILC (quadratic criterion-based ILC) was used for the tracking control of the reactor temperature. A time-varying reference trajectory for the reactor temperature that can meet the target conversion and purity of MAN was found through repeated simulations. A linear model for the design of the QILC was identified in the form of the output error (OE) model using experimental data.

To our knowledge, the present paper is the first contribution that applies an ILC technique to BD or RBD processes.

PROCESS DESCRIPTION

The RBD process produces MAN from methacrylic acid (MAA) and acrylic acid (AA). The reaction is described by the following two-stage elementary reversible reactions:

$$\mathrm{MAA}+\mathrm{AA} \underset{r_2}{\overset{r_1}{\rightleftharpoons}} \mathrm{Mxd}+\mathrm{AcH} \tag{1}$$

$$\mathrm{MAA}+\mathrm{Mxd} \underset{r_4}{\overset{r_3}{\rightleftharpoons}} \mathrm{MAN}+\mathrm{AcH} \tag{2}$$

where Mxd represents mixed anhydride. The overall reaction equation is

$$2\mathrm{MAA}+\mathrm{AA} \rightleftharpoons \mathrm{MAN}+2\mathrm{AcH} \tag{3}$$

The boiling points at 1 atm are 117.8, 155.1, 159.2, 205.0, and 209.9 °C for AcH, AA, MAA, MAN, and Mxd, respectively. If AcH, which has the lowest boiling point, can be removed continuously during the reaction, the equilibrium can be shifted to the right, enabling complete conversion to MAN.

A diagram of the RBD process is shown in Fig. 1. The process consists of a distillation column, a reactor vessel, a reflux drum, a product tank, and a vacuum pump system. The role of the distillation column is to continuously withdraw AcH. Because MAN undergoes thermal decomposition and polymerization at high temperature, the boiling temperature in the reactor should be maintained below a certain safety limit. For this, the column pressure is set at 20 torr at the top, and a packed column is employed to minimize the pressure increase at the bottom. The boiling point of MAN at 20 torr is 95 °C contrary to 205 °C at 1 bar.

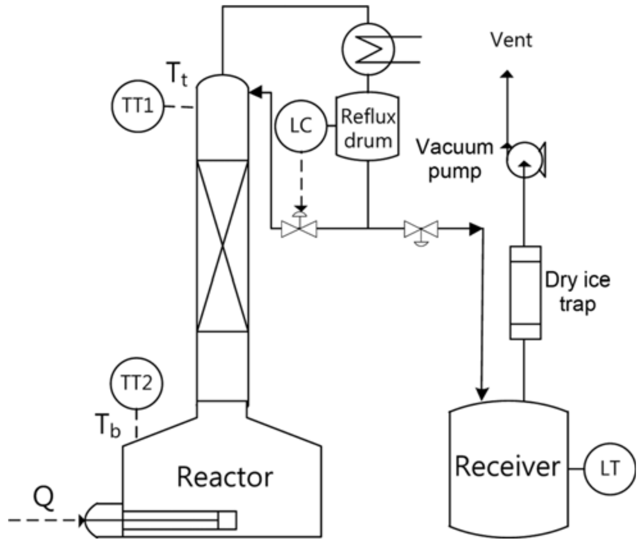

Fig. 1. The reactive batch distillation process for the production of MAN.

Table 1. Specifications of the RBD process

Distillation column	
Column type	Packed column
Packing type	Raschig Ring
Equivalent no. of stages	6
Section diameter	0.45 m
Height of packing	3.7 m
Packing size	0.015 m
Surface area of packing	350 m^2/m^3
Volume of the reactor vessel	4 m^3

The heat input to the reactor is used for the heat of reaction and the heat of vaporization for distillation. Excessive heat input causes local hot spots on the inner wall of the reactor and/or column flooding.

The numerical RBD process was constructed using the Batch Distillation module in ASPEN Plus^{TM}. Some primary specifications of the process are listed in Table 1. The values in Table 1 are actually from the pilot plant, where test experiments were conducted. The reaction rates, vapor-liquid equilibrium data, and other physical properties were provided by Samsung Cheil Industries Inc.

The performance of the RBD simulator using the ASPEN Plus^{TM} is demonstrated in Fig. 2. Estimated values by the simulator and measured values from the pilot plant in Samsung Cheil Industries, Inc. are compared for the time-dependent amount of AcH component withdrawn to the receiver (Fig. 2(a)), purity of MAN in the reactor vessel (Fig. 2(b)), temperatures (Fig. 2(c)) at the column top and bottom, and column pressure (Fig. 2(d)), respectively. AcH accumulation and MAN purity could be measured only once for

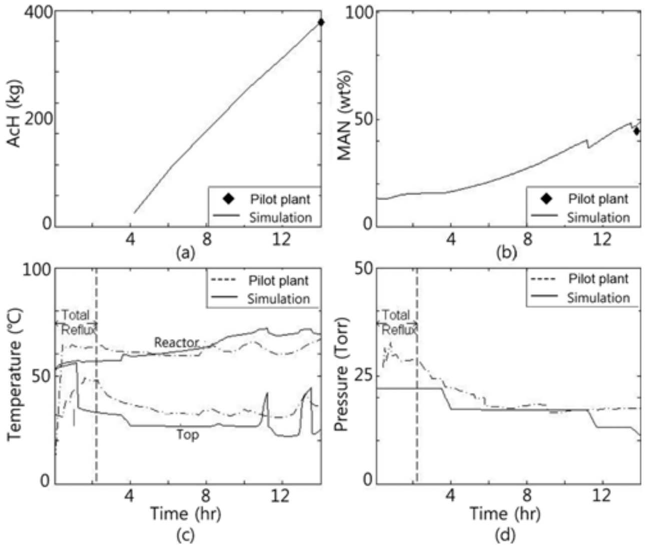

Fig. 2. Performance of the RBD simulator using the ASPEN Plus™. Simulation and pilot plant operation results are compared.

each, but they coincide with the simulation results very closely. The temperature and pressure profiles agree reasonably well except for some insigificant periods of time. Considering that all the reaction kinetic and thermodynamic parameters except for AcH were newly obtained through preliminary researches, the prediction of the process behaviors by the simulator is thought to be reasonble as a whole.

Fig. 3 displays typical trajectories for the selected process variables over the entire batch horizon. Shortly after the initiation of the reaction, AcH is separated in a highly pure state from the top where the top temperature is regulated at a temperature that is slightly above the boiling point of AcH at 20 torr. The mole fraction of MAN in the reactor vessel is increased up to 0.53. The reactor temperature represents the boiling point of the liquid mixture in the reactor vessel shown in Fig. 3.

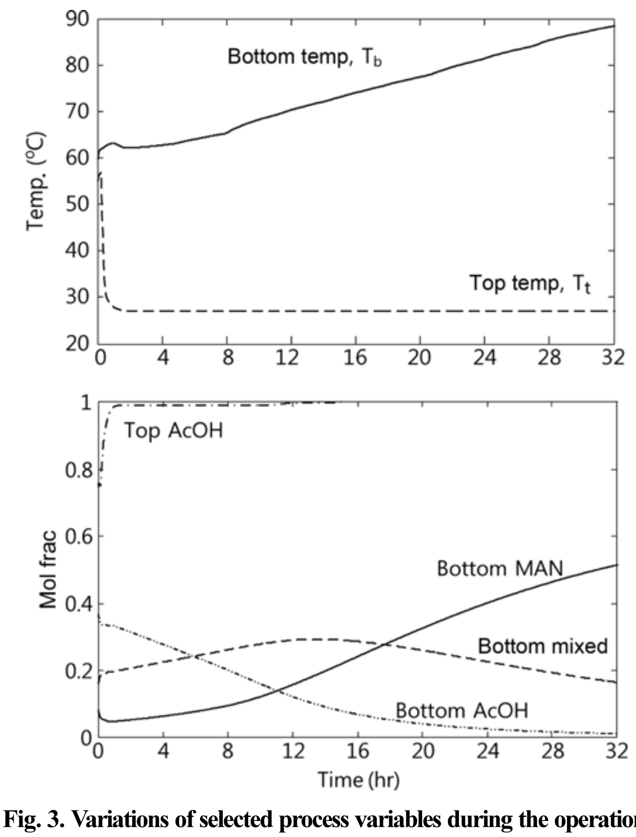

Fig. 3. Variations of selected process variables during the operation of the RBD process.

ITERATIVE LEARNING CONTROL

The RBD operation poses a problem to design two single-input single-output (SISO) controllers, one for the regulation of the top temperature and the other for the tracking of the bottom temperature. Regulation without a severe disturbance is normally easier than tracking, and satisfactory performance can be attained in many cases with PID control. Conversely, PID control may perform unsatisfactorily in tracking of a continuously varying reference trajectory. Control of the reactor temperature for the RBD process is an example of such cases. For improved tracking by exploiting the repetitive nature of the RBD operation, a QILC technique was adopted.

The QILC technique has been presented in different formulations [7]. The deterministic and constrained version of QILC was employed in this study since no severe disturbances enter the process during the operation while it is necessary to constrain the magnitude of the heat input. The algorithm is briefly described for the SISO case.

Consider a batch process with $u(t), y(t)\in\mathbb{R}$ as the input and output variables. The process can be modeled as a nonlinear static system between the input and output sequences defined over the batch horizon with N sampling times as follows:

$$\mathbf{y}=\mathbf{N}(\mathbf{u})+\mathbf{v} \tag{4}$$

where $\mathbf{u}\triangleq[u(0)\ \cdots\ u(N-1)]^T$ and $\mathbf{y}\triangleq[y(1)\ \cdots\ y(N)]^T$; $\mathbf{v}\triangleq[v(1)\ \cdots\ v(N)]^T$ represents the residual term.

Linearization of Eq. (4) around reference input and output trajectories produces

$$\mathbf{y}=\mathbf{G}\mathbf{u}+\mathbf{d} \tag{5}$$

where

$$\mathbf{u}\triangleq[u(0)\cdots u(N-1)]^T,\ \mathbf{y}\triangleq[y(1)\cdots y(N)]^T,$$
$$\mathbf{d}\triangleq\mathbf{y}_{ref}-(\partial\mathbf{N}/\partial\mathbf{u}^T|_{\mathbf{u}_{ref}})\mathbf{u}_{ref}+\text{higher-order terms}+\mathbf{v}$$

$$\mathbf{G}=\left.\frac{\partial\mathbf{N}}{\partial\mathbf{u}^T}\right|_{\mathbf{u}_{ref}}=\begin{bmatrix} g_{1,1} & 0 & \cdots & 0 \\ g_{1,2} & g_{2,1} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ g_{1,N} & g_{2,N-1} & \cdots & g_{N,1} \end{bmatrix} \tag{6}$$

As can be understood from the linearization procedure, **u** and **y** in Eq. (5) need not be deviation variables. True physical variables can be used for them. In Eq. (6), $g_{i,j}$ represents the j^{th} pulse response coefficient to a unit pulse input at i.

Let $\mathbf{r}\triangleq[r(1)\ \cdots\ r(N)]^T$ and $\mathbf{e}\triangleq\mathbf{r}-\mathbf{y}$ be the sequences of the reference trajectory and the output error, respectively. Also let $\Delta\mathbf{u}_{k+1}\triangleq\mathbf{u}_{k+1}-\mathbf{u}_k$ and the subscript k denotes the batch index. The constrained QILC solves

$$\min_{\Delta\mathbf{u}_{k+1}}\frac{1}{2}\{\mathbf{e}_{k+1|k}^T\mathbf{Q}\mathbf{e}_{k+1|k}+\Delta\mathbf{u}_{k+1}^T\mathbf{R}\Delta\mathbf{u}_{k+1}\} \tag{7}$$

subject to $\mathbf{e}_{k+1|k}=\mathbf{e}_k-\mathbf{G}\Delta\mathbf{u}_{k+1}$

$\Delta\mathbf{u}_{min}\le\Delta\mathbf{u}_{k+1}\le\Delta\mathbf{u}_{max}$

$\mathbf{u}_{min}\le\mathbf{u}_{k+1}\le\mathbf{u}_{max}$

after the kth batch run, and $\mathbf{u}_{k+1}=\mathbf{u}_k+\Delta\mathbf{u}_{k+1}$ is implemented for the k+1th batch run. Eq. (7) is a quadratic programming problem, and the global minimum can be found in a reliable and efficient way. When the inequality constraints are absent, $\mathbf{u}_{k+1}$ can be obtained in the analytic form as:

$$\mathbf{u}_{k+1}=\mathbf{u}_k-(\mathbf{G}^T\mathbf{Q}\mathbf{G}+\mathbf{R})^{-1}\mathbf{G}^T\mathbf{Q}\mathbf{e}_k \tag{8}$$

QILC can attain asymptotically perfect tracking for unknown but run-invariant **d** and model error for any choice of **Q** and **R**, unless QILC is divergent. When **R**/**Q** is large, QILC becomes more robust against model error but the convergence rate is low and vice versa [6].

CONTROLLER IMPLEMENTATION

The RBD process has three variables to control, which include the top and bottom temperatures, and the column pressure. The column pressure can be independently controlled by manipulating the vacuum pump. In the Batch Distillation module of ASPEN Plus™, the column pressure is an independent variable that can be set at an arbitrary constant value; thus control of the top and bottom temperatures is considered in this study.

Fig. 4 shows the overall control structure of the RBD process, in which the top and bottom temperatures are controlled by two SISO controllers, PID and QILC, with the reflux ratio and the reactor heat input as the MVs, respectively.

1. PID Controller, TC1

The top temperature, T_t, cannot be decreased below the boiling point of AcH, 27 °C at 20 torr. The set point of T_t was given at 28 °C for the temperature error to be able to move in positive and negative directions. The steady state relationship between the reflux ratio and T_t is highly nonlinear near the set point, as is drawn in Fig. 5. For linearization, T_t is transformed to $z=\ln((T_t-26.9)/1.1)$ and fed to TC1.

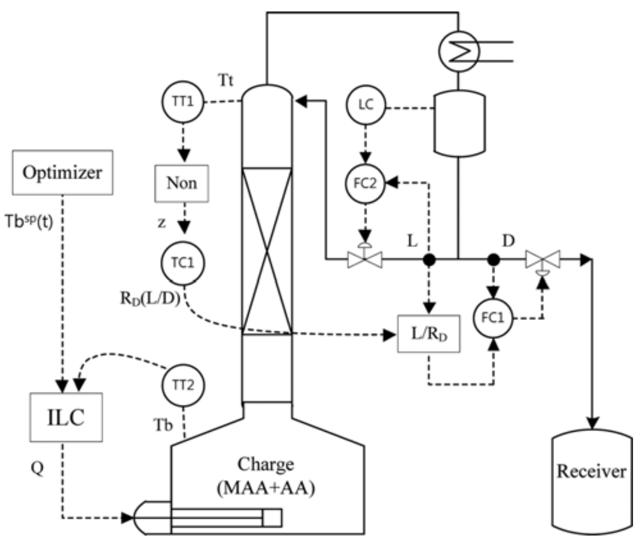

Fig. 4. Control scheme of the RBD process.

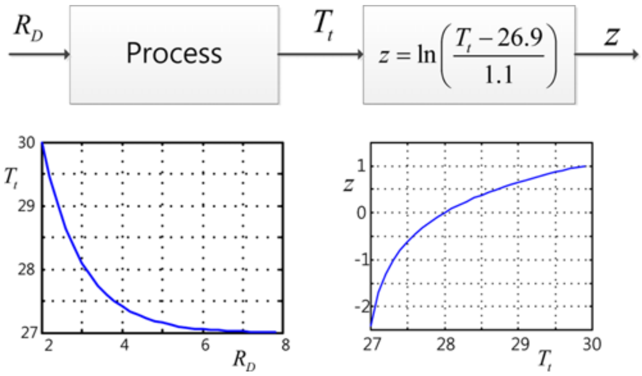

Fig. 5. Linearization of the top temperature measurement.

TC1 sends out the reflux ratio R_D and the corresponding distillate flow rate, D, is computed, as shown in Fig. 4. TC1 was tuned by using an experimentally determined first-order plus dead time model and the IAE criterion [8].

2. Model Identification and QILC

For QILC construction, a model between the reactor heat input, u (=Q), and the bottom temperature, y (=T_b), should be available in the form of the **G** matrix. For this, identification experiments were conducted by superimposing a binary sequence on the nominal Q(t) trajectory, and the T_b(t) response was taken. Using $\bar{u}(t)$ and $\bar{y}(t)$ as the input and output deviations from their nominal trajectories, the input-output relationship was identified as the following time-invariant output-error model [9].

$$\bar{y}(t)=\frac{B(q^{-1})}{A(q^{-1})}\bar{u}(t-d)+e(t) \tag{9}$$

'ident.m' in MATLAB was used for identification. Consequently, A, B, and d were estimated as

$A(q^{-1})=1-3.3492q^{-1}+4.0569q^{-2}-2.0660q^{-3}+0.3584q^{-4}$
$B(q^{-1})=0.0303q^{-1}-0.0857q^{-2}-0.0805q^{-3}-0.0251q^{-4}$
$d=3$

for the sampling time of 1.9 min. The coefficients of long division of $q^{-3}B(q^{-1})/A(q^{-1})$ correspond to the unit pulse response coefficients of the process, with which **G** was constructed.

QILC determines the input sequence for the next batch on the basis of the open-loop output prediction. Hence, the open-loop prediction performance is important for the identified model, which is the reason for choosing the output-error model. Fig. 6 shows an input-output data set used for the identification, and Fig. 7 shows the com-

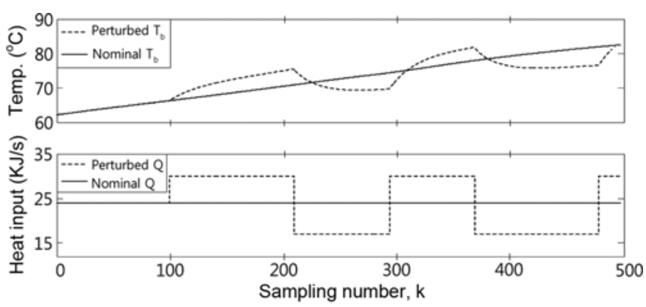

Fig. 6. Input and output data for identification.

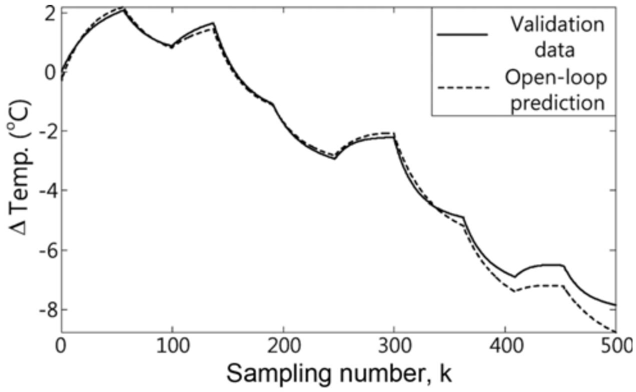

Fig. 7. Validation of the identified model.

parison of the open-loop output prediction by the identified model with a validation data set. Interestingly, a quite accurate open-loop model could be found over the control horizon for the concerned process.

The sampling time was chosen as 1.9 min throughout the study and QILC was applied over the first half of the batch horizon, which is equivalent to 500 sampling instances. The input change was, however, made only ten times over the control horizon, which corresponds to once every 50 sampling instances or every 95 min. The output weight matrix **Q** was set as **I**, and the input weight matrix **R** was given as λ**I** with λ=0.1. Constraints were imposed only on the heat input u_{min}=0 with and u_{max}=28 kJ/sec, not on the input change.

RESULTS AND DISCUSSION

Constrained QILC was first applied to the identified linear model and the tuning factor λ was adjusted through repeated simulations such that the heat input Q(t) does not excessively violate the constraints. Fig. 8 shows the results for selected batches over four consecutive batch runs. The output converges very closely to the reference trajectory at the fourth run while the heat input hits the constraint one time.

Constrained QILC with l tuned as above was applied to the simulated RBD process, and the results over four batch runs are in Fig. 9. The reference temperature trajectory in Fig. 9 was developed by Samsung Cheil Industries, Inc. such that the MAN yield after a batch

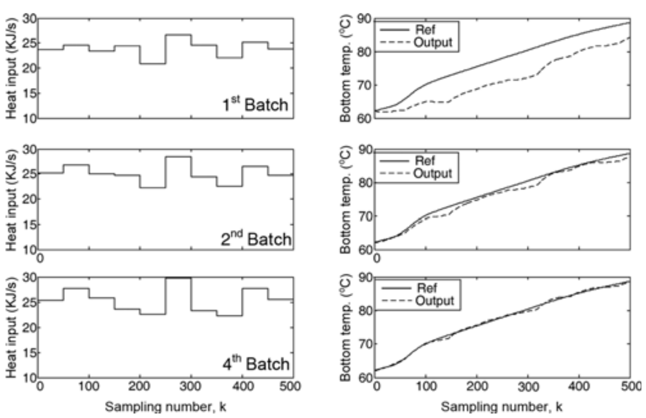

Fig. 8. Result of QILC for the identified linear model.

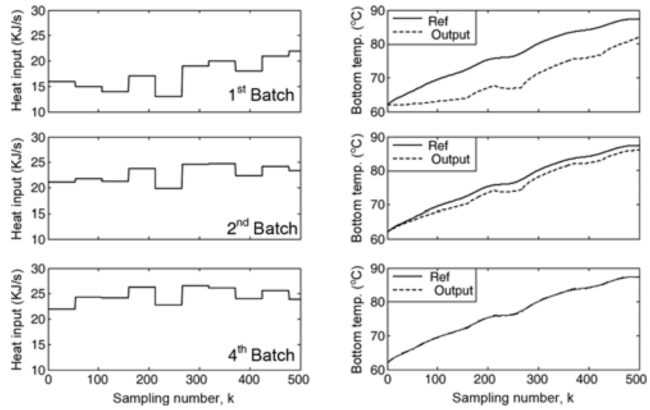

Fig. 9. The results of QILC for the simulated reactive batch distillation process using ASPEN Plus™.

operation is high enough. The bottom temperature converges to the reference trajectory as the batch number increases. Overall output profiles looking over the consecutive batches look similar to those in Fig. 8. The heat input variation during a batch run, however, appears milder than in Fig. 8 and does not hit the constraint.

For comparison, PID control was attempted instead of QILC for tracking the bottom temperature, and the representative result is shown in Fig. 10. The PID parameters were determined by applying the IAE criterion to the identified model. The tracking performance is not as good as that of QILC but is acceptable. The trouble is that the highly oscillatory input signal often far exceeds u_{max}, especially during an early period. Another notable consequence is that the mole fraction of MAN at k=500 under PID control was found to be 0.316, while the mole fraction under QILC was 0.344. It seems that the negative deviation of the reactor temperature from the reference trajectory during the initial period causes the decrease in the yield of MAN.

CONCLUSIONS

A control system for an RBD process that produces MAN was proposed, and its performance was investigated. The major role of batch distillation is to continuously remove AcH so that the yield of MAN in the bottom reactor vessel could be maximized by shifting the equilibrium conversion. The control system consists of two independent control loops for the top and bottom temperatures, respec-

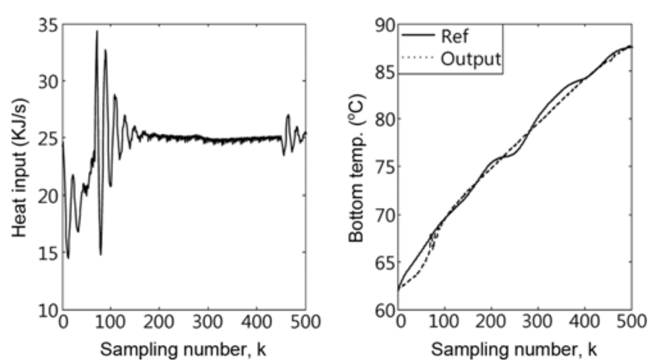

Fig. 10. Results of PID of reactive batch distillation.

tively. Regulation is required for the top temperature, and a nonlinear PID controller was used for this purpose. For the bottom temperature, tracking a specified profile is required, and constrained QILC was employed to achieve this goal. The proposed control system design showed satisfactory performance.

The RBD model programmed using Batch Distillation in ASPEN Plus™ was found to closely represent the behavior of the real pilot process in Samsung Cheil Industries Inc. In this respect, the proposed QILC method can be a practical tool for the RBD process. For real process applications, QILC needs to be equipped with real-time feedback control [1,10]. Unfortunately, the present version of the Batch Distillation module in ASPEN Plus™ does not allow the implementation of a customized real-time feedback controller except the embedded PID controller, and QILC combined with real-time feedback control could not be investigated in this study.

ACKNOWLEDGEMENTS

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