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Operability and Proportional Integral Control of Reactive Distillation Configurations

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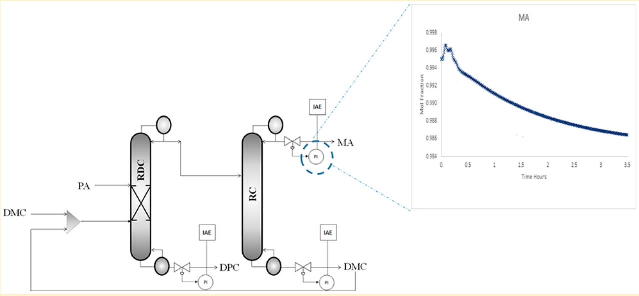
Supporting Information

ABSTRACT: This work addresses two key issues in the design of control systems based on proportional integral (PI) lineal controllers for intensified reactive distillation configurations to produce diphenyl carbonate: (i) the practical controllability of the process that inherently leads to the setting of control configurations and (ii) the tuning of corresponding PI controllers in a systematic framework. For the first issue, through the relative gain array the appropriate control loops are established, and an operability index is proposed as a complement to establish the practical feasibility of control loops in complex and highly sensitive systems. For the second issue, a technique based on stable pole assignment is applied, where explicit tuning relationships enable the simultaneous adjustment of all control loops through only one parameter. The performance of the control systems is illustrated through simulations which show that resulting control configurations are effective, and that PI controllers can be tuned in a practical and systematic framework.



1. INTRODUCTION

Developments in the chemical industry have provided a wide range of products that have improved modern life; in addition, there has been a demand boom during the past decades due to the low-cost energy coming from fossil materials such as petroleum.[1] However, the end of the low-cost energy era is looming due to the limited sources and to the huge and fast-growing demand for energy from developing countries. Moreover, diminishing greenhouse gas emissions is an increasingly worldwide necessity, which is directly influenced by the consumption of fossil fuel energy sources.

The chemical industry is strongly affected by the cost of energy sources, and enhancing the energy usage efficiency is of paramount importance. In this light, the reactive distillation column (RDC) is an excellent example of process integration and innovation, since it combines two conventional processes of reaction and separation into one single equipment to obtain significant savings on energy consumption and also in capital investments.[2]

In addition to savings, RDC enables improvement of product selectivity due to a rapid depletion of reactants or removal of products from the reaction zone.[3] Nevertheless, RDC also has disadvantages such as the existence of multiple stationary states and high sensitivity to fluctuations of operational variables due to the combination of separation and chemical reaction. Therefore, special attention must be paid to the design of its control system.[4] It can be difficult to control a reactive distillation column that operates in this orderly fashion. The problem is the need to feed in exactly enough of the reactants and the simultaneous reaction with the separation. If the material and energy balances are not absolutely perfect in the column, it will not be possible to conserve product purities.[5]

A first step in the design of a control system is the determination of the feasibility of controlling the process with the available control inputs. Recalling the words of Georgakis et al.,[4] "it is necessary to determine the inherent ability of the process to move from one steady state to another and to reject any of the expected disturbances in a timely fashion with the limited control action available."

Several approaches have been followed to determine the feasibility of controlling the process, such as the proper definition of controllability[6] and the singular values decomposition (SVD) technique.[7] The approach based on a controllability definition is hard to apply in distillation columns because they are modeled by many highly nonlinear equations. By using an approximate linear model of the process, the SVD technique seems to be appropriate, and it has been widely used;[8,9] however, this

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technique only provides a relative viewpoint since measuring parameters of controllability for certain process must be compared with the ones of similar systems of proved control feasibility. Aiming toward a practical sense of controllability called *process operability*, Georgakis et al.[10,11] have proposed several indices that correlate the workspace of available inputs with the workspace of achievable outputs and desired outputs, in such a way that the feasibility of obtaining the desired output ranges with the available input ranges is thoroughly determined. Although the characterization of such feasibility does not depend on the model linearity and controller type, a reduced model is suggested for a high-dimensional and nonlinear process. In distillation columns, a reduced model means an approximate linear model identified from simulated trajectories coming from a rigorous model.

Despite its importance, controllability assessment is frequently overlooked when designing control systems for distillation columns. In other situations, it seems to be covered intrinsically by setting up a likely effective control configuration based on the relative gain array (RGA) method,[12] which is straightforwardly derived from the steady-state gains of an approximate linear model of the process.[13] Nevertheless, the information obtained from RGA does not guarantee control feasibility; for example, if a control loop does not work, it is not possible to know whether the lack of controllability is due to an unsuitable controller pairing, or an inappropriate controller tuning.

Once the controllability and the control configuration have been established, the next step consists of constructing and implementing the process controllers. For distillation columns, the linear proportional−integral−derivative (PID) controller is widely used, and its implementation depends on tuning it effectively. It is worth recalling that, although distillation processes are nonlinear systems, the PID controller is enough to attain good control performance even in processes with heat integration or chemical reaction.[14] Thus, the control system performance depends on the tuning of controllers.

The most commonly used tuning techniques are the Ziegler−Nichols[15] and Ling−Luyben[16] since they are based on characteristic parameters coming up from linear models. For high-dimensional and nonlinear distillation systems, these approximate models are identified through the process reaction curve method.[17] However, the resulting values for the gains of each controller in the distillation control system are implemented not simultaneously, but sequentially: first, one controller (typically the one closer to the column top) is implemented with the gain calculated from a tuning technique while the other outputs remain in open loop; then, this closed-loop performance is tested while the tuning parameters are readjusted. Next, the second controller is implemented, and the control system performance is tested while readjusting the tuning parameters. In this sense, tuning PID controllers in distillation systems through above-mentioned techniques require neat extensive trial-and-error evaluations. In another way, following an approach based on stable pole assignment, Zavala-Guzmán et al.[18] achieved the simultaneous and systematic tuning of the PI controllers for a dividing-wall distillation column (DWC). The approach relies on a first-order linear approximation of the behavior of each input−output of control loops, and resulting tuning relationships are easily applicable, ultimately leaving only one parameter to be adjusted for all the control loops.

Numerous papers have been written on and patents have been granted in the area of reactive distillation. Most of these works have treated subjects such as steady-state design of reactive distillation columns. Conceptual approximate design approaches are emphasized, but treatment of rigorous design approaches that use commercial simulators and address the issue of dynamics and control structure development are not covered. Nevertheless, there are works that address the problem of control in reactive distillation columns. The first work reported on these topics is from Roat et al.[19] They confirmed the inadequacies of conventional linear multiloop controllers with input−output pairings established on steady state interaction measures, and they highlighted the need for more advanced controllers designed on the basis of rigorous dynamic models. Then Kumar and Daoutidis[20] show a detailed dynamic model that resulted and was used for the design of a nonlinear controller for a column with a kinetically limited reversible reaction. The dynamic behaviors of batch reactive distillation columns have also been studied in the scheme of optimal control by Sørensen et al.,[21] and in nonlinear model predictive control applying reduced order models by Balasubramhanya and Doyle.[22] In recent times, Luyben and Yu[5] presented a comprehensive treatment of both steady-state design and dynamic control of reactive distillation systems using rigorous nonlinear models. Despite all these reported works, none of them shows a design control system for reactive distillation columns in a systematic, simple, and quick way.

This work aims to design control systems for reactive distillation columns in a systematic way, and the production of diphenyl carbonate (DPC) is considered a challenging case study. We have attempted to deal with issues in the reactive distillation and control challenges in a simple and prompt manner, this being a preliminary control study. In this framework, the first problem of establishing an effective control configuration is addressed by complementing the RGA technique with a parameter that measures the change in a certain control input to cause a change in a certain control output, something like just evaluating a single point of a space that would be generated by following Georgakis' approach. Next, considering linear PI controllers, the problem of tuning corresponding gains is addressed by exploring the direct application of tuning relationships that resulted from the stable pole assignment approach in Zavala-Guzmán et al.[18] In the final part, via simulation, the performance of control systems is discussed.

2. CASE STUDY

DPC is an important precursor in the production of polycarbonate, which has several commercial applications thanks to its electrical, mechanical, optical, and heat-resistance properties.[23] Tuinstra and Rand[24] proposed a very promising route of obtaining DPC via RDC. In this work, three reactive distillation configurations are considered: a conventional one, a thermally coupled configuration, and a vapor recompression and thermally coupled reactive distillation. Like any other process, in order to maintain maximum performance, RDC to produce DPC must be controlled to reject operative disturbances, such as changes in the raw material quality or temperature or flow of inlet stream. Therefore, it is highly important to establish an effective control system for the process.

2.1. Reactive Distillation Processes and their Control Systems.

The conventional production of diphenyl carbonate (DPC) considers a stirred tank reactor where an esterification reaction with dimethyl carbonate (DMC) and phenyl acetate (PA) takes place:

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$$\mathrm{DMC} + \mathrm{PA} \rightleftarrows \mathrm{MPC} + \mathrm{MA} \tag{1}$$

$$\mathrm{MPC} + \mathrm{PA} \rightleftarrows \mathrm{DPC} + \mathrm{MA} \tag{2}$$

$$2\mathrm{MPC} \rightleftarrows \mathrm{DPC} + \mathrm{DMC} \tag{3}$$

The overall reaction is

$$\mathrm{DMC} + 2\mathrm{PA} \rightleftarrows \mathrm{DPC} + 2\mathrm{MA} \tag{4}$$

Since the reaction is reversible, with methyl acetate (MA) as a byproduct, a downstream step of DPC purification is carried out through two distillation columns; in the first one DPC is recovered, and in the second one DMC is separated from MA and sent back to the first column.

The rate expressions for the reversible reactions of eqs 1–3 are eqs 5–7.

$$r_1 = k_1 C_{\mathrm{PA}} C_{\mathrm{DMC}} - k_{-1} C_{\mathrm{MPC}} C_{\mathrm{MA}} \tag{5}$$

$$r_2 = k_2 C_{\mathrm{PA}} C_{\mathrm{MPC}} - k_{-2} C_{\mathrm{DPC}} C_{\mathrm{MA}} \tag{6}$$

$$r_3 = k_3 C_{\mathrm{MPC}}{}^2 - k_{-3} C_{\mathrm{DPC}} C_{\mathrm{DMC}} \tag{7}$$

The reaction rate coefficients of three reversible reactions are related as follows:

$$\frac{k_2}{k_{-2}} = \frac{k_1}{k_{-1}} \frac{k_3}{k_{-3}} \tag{8}$$

Cheng et al.[23] show the data for the utilized kinetic parameters that fit the Arrhenius equation. Table S1 (Supporting Information) shows the numerical values of the pre-exponential factor, k_0, and the activation energy, E_a, of each reaction rate coefficient.

Azeotropes are not present in the components; however, there is a large difference in the boiling points. An ideal thermodynamic model would adequately represent the equilibrium vapor–liquid system of DMC and PA, as experimentally demonstrated by Yao.[25]

2.2. Reactive Distillation Configurations. The conventional system described above is limited by the reaction reversibility. Therefore, it has been rearranged to a reactive distillation configuration in order to move the equilibrium toward products, and inherently to reduce the energy consumption in the column by using the heat released from the reaction. However, for the reaction system in eqs 1–3, the first reaction is slightly exothermic. Thus, its heat of reaction is used to promote vaporization of low boiling point components in the reactive stages where this reaction takes place rather than supplying heat to another distillation column or any part of the process.[19] This work considers the following three reactive distillation configurations:

1. The conventional reactive distillation (CRD) configuration (Figure 1) consists of two distillation columns. In the first one, which is the reactive column, the reaction is carried out with a PA conversion of 99%, and the product stream contains a DPC molar purity of 99.5%. In the second one, which is the recovery column, the remaining reactant DMC and the byproduct MA are separated through a conventional distillation process. The recovered DMC is recycled to the reactive column.

2. The thermally coupled reactive distillation (TCRD) configuration (Figure 2) is similar to CRD; however, the condenser of the reactive column is replaced by coupling the top stream with side stream in the recovery column. The enthalpy of the vapor stream leaving the reactive column is used in the second column; as a consequence, one condenser is eliminated. Also, the remixing effect is eliminated in the first column. The elimination of the remixing effect in reactive distillation columns results in energy savings.[26]

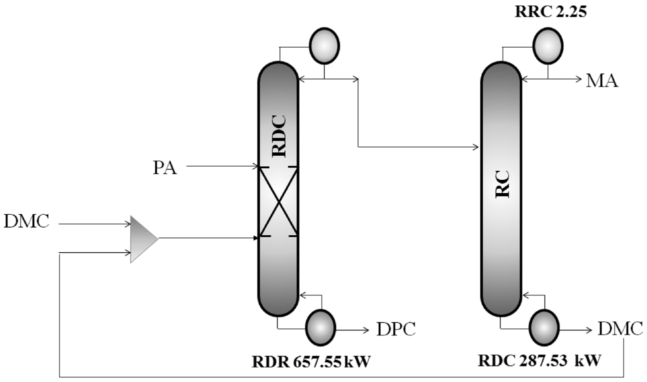


Figure 1. Conventional reactive distillation (CRD) configuration for the synthesis of DPC.

3. The vapor recompression and thermally coupled reactive distillation (VTCR) configuration (Figure 3) is derived from TCRD. The liquids and vapors were directly interchanged between the reactive column and the recovery column. This heat integration is aimed toward providing additional energy savings in the reboiler of recovery column.

These three configurations are adopted from Contreras-Zarazúa et al.,[27] where a more detailed explanation of the designs can be found. Such results exhibit the highest energy savings among a set of optimized configurations. The parameters resulting from each process can be observed in the Supporting Information (Table S2). The study was carried out in a rigorous simulation framework supported by the software Aspen Plus. The design parameters were exported to Aspen Plus Dynamics in order to carry out the dynamics analysis.

3. PROBLEMS ON THE DESIGN OF CONTROL SYSTEMS

The three reactive distillation configurations share a common goal, which is to maintain three key compositions in product streams: (i) DPC in the bottom stream of the reactive column, (ii) DMC in the bottom of the recovery column, and (iii) MA in the top stream of the recovery column. Keeping a high DPC composition is mandatory because of product quality requirements,[27] a high MA composition is convenient because it means an almost complete recovery of DMC, and in turn a high DMC composition means an almost pure DMC recycle. These requirements create a tight control problem of composition in product streams.

The systems CRD (Figure 1) and TCRD (Figure 2) have the following control input choices: (i) reboiler duty of the reactive column (RDR), (ii) reboiler duty of the recovery column (RDC), and (iii) reflux ratio of the recovery column (RRC). The system VTCR (Figure 3) has the following control input choices: (i) RDR, (ii) work duty in the compressor (WDC), and (iii) RRC. The difference in the latter configuration is due to the use of process-to-process heat integration. The RDC depends on the heat supplied by the heat exchanger in which the input comes from the compressor (QVR). Therefore, RDC cannot be set as the control input. In this work, WDC is taken as an independent control input to deal with changes in the purity of DMC.

In this way, the reactive distillation configurations are systems of three control inputs–three control outputs. For the systems CRD and TCRD, the input and output sets are

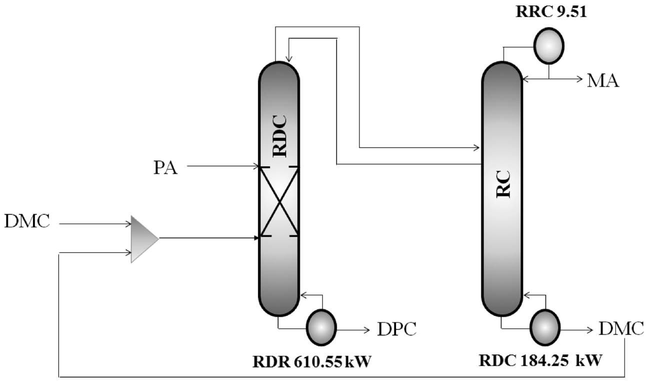


Figure 2. Thermally coupled reactive distillation (TCRD) configuration for the synthesis of DPC.

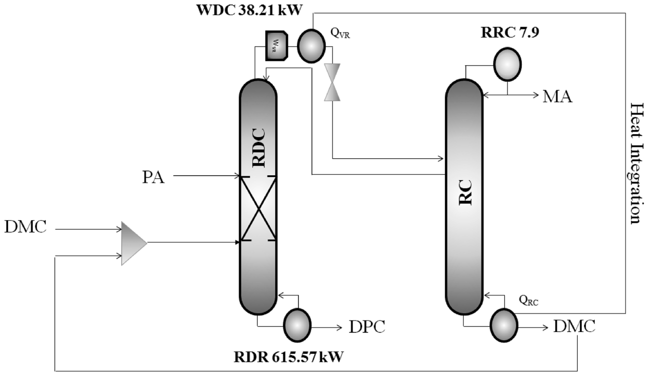


Figure 3. Vapor recompression and thermally coupled reactive distillation (VTCR) configuration for the synthesis of DPC.

$$\begin{aligned}(u_1, u_2, u_3) &= (\text{RDR, RDC, RRC}),\\ (y_1, y_2, y_3) &= (\text{DPC, DMC, MA})\end{aligned} \tag{9}$$

while the ones for the system VTCR are

$$\begin{aligned}(u_1, u_2, u_3) &= (\text{RDR, WDC, RRC}),\\ (y_1, y_2, y_3) &= (\text{DPC, DMC, MA})\end{aligned} \tag{10}$$

Therefore, the establishment of the control configuration for each reactive distillation system implies the assessment of nine possible control loops.

On the second problem of controller construction, trivially for every input−output pair, a conventional linear PI controller is considered:

$$u(t) = \bar{u} + k_{\text{P}}(y(t) - \bar{y}) + \frac{k_{\text{P}}}{\tau_{\text{I}}}\int_0^t (y(\theta) - \bar{y})\,\mathrm{d}\theta \tag{11}$$

where u = RDR or RDC or RRC or WDC, and y = DPC or DMC or MA, according to the established control loops. Here k_{P} is the proportional gain, whereas τ_{I} is the integral time. Next, the subsequent third problem corresponds to determining the values of the proportional gain and integral time of controllers. It is worth recalling that this task involves a great operative effort, as described above, and the challenge of this work is to carry it out in a systematic way.

The performance of the control system in distillation columns is hard to characterize, and this is typically assessed by simulation of scenarios of servo-control problems. Therefore, this fourth problem will be tackled in this typical way, and with a scenario of regulatory control problem to assess disturbance rejection.

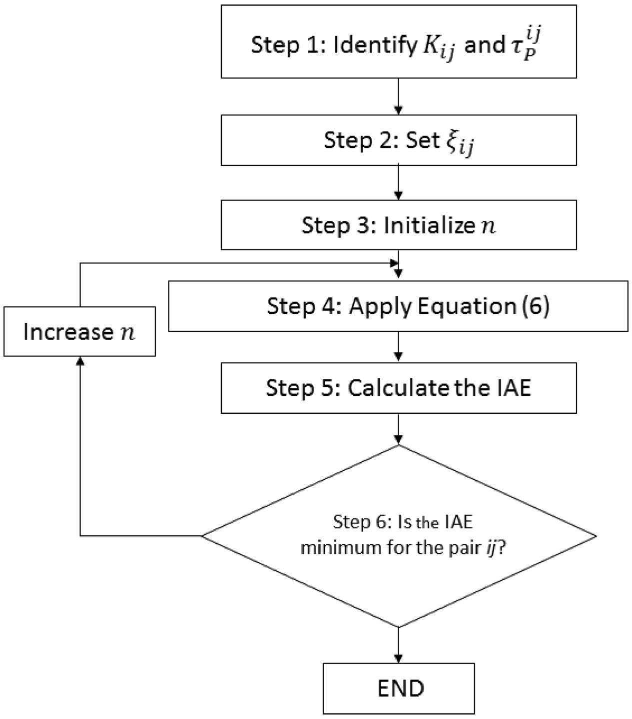


Figure 4. Flowchart for tuning parameters of controllers.

Then, the control system design relies on establishing a control configuration for each reactive distillation configuration, tuning the gains of the PI controllers, and verifying the effectiveness of the established control systems.

It is worth saying that continuous−instantaneous measurements of composition are assumed in this work, but in practice this kind of variable is measured with delay, and possibly in discrete form as well, depending on the available infrastructure to analyze the components involved in the process. Toward an implementation work, it can be resorted to the use of online continuous−instantaneous measurements of temperature, but offsets between the desired composition and the current one will likely result.[26] Therefore, other adjustments must be considered as the implementation of an observer.[28] If discrete−delayed composition measurements were considered, a corresponding tuning technique as in Zavala-Guzmán et al.[18] can be followed.

4. CONTROL SYSTEM CONFIGURATION

Following the heuristic approach in which a control input is paired with the nearest control output, an immediate control input−control output pairing that can be set up is as follows: (RDC, DMC), (RDR, DPC), and (RRC, MA). However, the other six configurations could be suitable or even better, so they are worthy of being assessed. By following the RGA approach is the typical way to carry out this task. In this work, this task is complemented through a parameter called the operability index and their results are easy to interpret.

4.1. Relative Gain Array Technique. The determination of a control configuration for MIMO systems is typically performed via the relative gain array (RGA) technique, particularly in distillation systems. Although it is well-known and well-described in any process control textbook, here it is worth recalling that, in relation to a specific input−output pair (u_i, y_j) among the ones that can be formed by the control inputs

Table 1. Results of Relative Gain Array (RGA) Analysis[a]

	MA	DMC	DPC
CRD			
RDR	1.98809	−0.38520	**3.76887**
RDC	−2.38366	**1.61911**	−2.60721
RRC	**1.39556**	−0.23390	−0.16165
TCRD	MA	DMC	DPC
RDR	−0.00020	0.27270	**0.72750**
RDC	−0.06140	**0.77990**	0.28150
RRC	**1.06160**	−0.05260	−0.0090
VTCR	MA	DMC	DPC
RDR	0.5671	−0.0032	**0.90712**
WDC	−0.0034	**1.0082**	−0.0012
RRC	**0.93431**	−0.0034	0.0764

[a] RDR, reboiler duty reactive column; RDC, reboiler duty recovery column; RRC, reflux ratio recovery column; WDC, work duty in the compressor.

Table 2. Results of Operability Indexes Analysis[a]

	MA	DMC	DPC
CRD			
RDR	1.75254	−2.87318	**0.40030**
RDC	−3.74942	**4.42030**	0.26092
RRC	**5.31855**	−11.90028	−0.37774
TCRD	MA	DMC	DPC
RDR	3.52675	−9.95489	**0.13600**
RDC	−68.67198	**27.55065**	1.66849
RRC	**4.72352**	−26.56499	−0.39713
VTCR	MA	DMC	DPC
RDR	−3.3687	−5.6781	**0.7678**
WDC	−54.8954	**12.5672**	−15.8320
RRC	**3.4456**	−13.4567	−0.6784

[a] RDR, reboiler duty reactive column; RDC, reboiler duty recovery column; RRC, reflux ratio recovery column; WDC, work duty in the compressor.

and control outputs in a certain system, the relative gain (λ_{ij}) is the ratio of two steady-state gains: the open-loop gain (i.e., all u_i are open) divided by the closed-loop gain (i.e., all u_i are open while the rest $u_{k\neq i}$ are closed with perfect control). Then, λ_{ij} is a measure of the effect on the input−output pair when the other inputs were to drive the other outputs to control the system. If λ_{ij} is greater than 1, it means that the effect of other loops is opposite to the main effect of u_i over y_j. If λ_{ij} is between 0 and 1, the effect of other loops is in the same direction as the main effect of u_i over y_j. Then, it is advised to choose control input−control output pairs whose relative gains are close to 1, which means that u_i can control y_j without interference from other control loops.

The RGA is an easy-to-apply technique since just static gains of the outputs with respect to the inputs are required, and these parameters can be identified through the reaction curve technique[17] when a process model is not available or is high-dimeansional and nonlinear, as in most times in complex distillation processes.

4.2. Operability Index.

The feasibility of a control input to modify a given control output can be visualized through its corresponding static gain; however, this information is not sufficient to determine whether the control output can effectively reach a desired point. In other words, although the control input can drive the control output, the required values in the control input may be unfeasible due to the process constraints involved in the control input span. In addition, if there is more than one choice of control input, and more than one control output, the matter turns into which control input requires less effort to drive a control output; in this way, the control input−control output pairing results in a combinatorial problem.

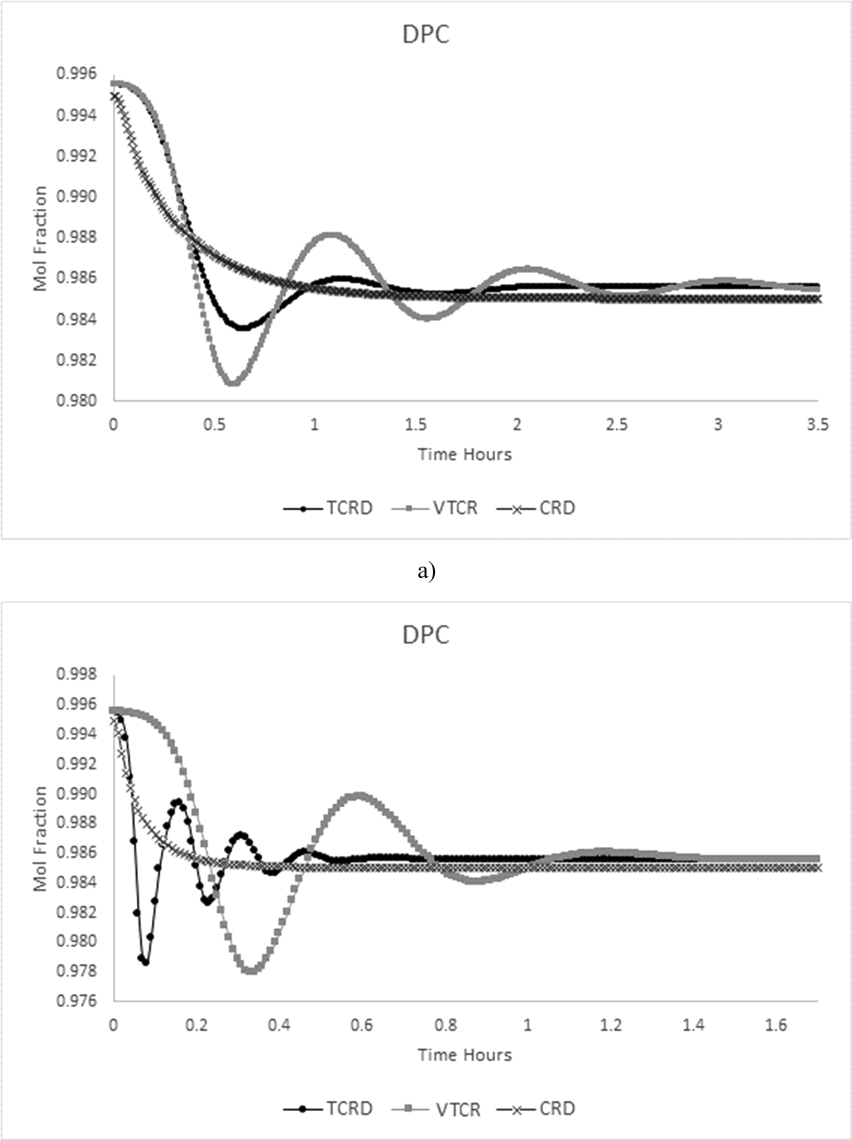
An estimation, in terms of percentage with respect to nominal values, of the change that must be made in certain control input (u_i) to drive a unit change in certain control output (y_j) is given by

$$\mathrm{OI}_{ij} = \frac{\bar{y}_j}{\bar{u}_i}\frac{1}{K_{ij}} \tag{12}$$

where $\bar{u}_i$ is the nominal value of u_i and $\bar{y}_j$ is the one of y_j; in turn, K_{ij} is the static gain of the control input−control output pair (u_i, y_j). Although K_{ij} can be accurately calculated through the linearization of the process model and a Laplace transformation to obtain the transfer function of the pair (u_i, y_j), this work uses the reaction curve method to calculate K_{ij}, which is suitable for high-dimensional and nonlinear models.[17,18] Although K_{ij} does

Table 3. Tuning Parameters and Controller Gains for Equal *n*

	k_P	τ_P (h)	ξ	n	k_C	τ_I (h)
CRD						
(RDR, DPC)	0.5772	2.3837		10	32.9183	0.3205
(RDC, DMC)	0.6996	3.2337	0.8412	10	27.1584	0.4348
(RRC, MA)	0.2392	12.2749		10	79.4328	1.6503
TCRD	k_P	τ_P (h)	ξ	n	k_C	τ_I (h)
(RDR, DPC)	4.5135	1.7099		10	4.2095	0.2299
(RDC, DMC)	0.2243	2.6962	0.8412	10	84.7180	0.3625
(RRC, MA)	0.2905	4.2987		10	65.3856	0.5779
VTCR	k_P	τ_P (h)	ξ	n	k_C	τ_I (h)
(RDR, DPC)	4.4456	1.4587		10	4.1721	0.2136
(WDC, DMC)	0.3213	4.4123	0.8412	10	54.9965	0.5835
(RRC, MA)	0.2235	2.9421		10	79.6781	0.4021



a)

b)

Figure 5. (a) Dynamic responses with the same n value in the DPC loops. (b) Dynamic responses with different n values in the DPC loops.

not consider the effect of other inputs, but only that of u_i, if there were other factors with an adverse effect, the OI_{ij} value could be regarded as the smallest change of u_i that yields the unit change of y_j. Therefore, if the OI_{ij} values for different control input−control output pairs were compared, the pair with the lowest value of OI_{ij} must be chosen. Finally, the comparison of the OI_{ij} values is visualized by arranging them into a matrix where the rows correspond to the inputs while the columns correspond to the outputs.

It is worth making notice that OI_{ij} can be seen as a paramenter of the sensitivity of y_j with respect to u_i that, in addition to K_{ij} by itself, takes into account the scale of the system through $\overline{u}_i$ and $\overline{y}_j$.

5. TUNING THROUGH POLE ASSIGNMENT

The methodology of tuning through pole assignment applied in this work was developed in the work of Zavala-Guzmán et al.[18] Therefore, once the control configuration has been set, on the tuning of linear PI controllers for each control input−control output pair (u_i, y_j)

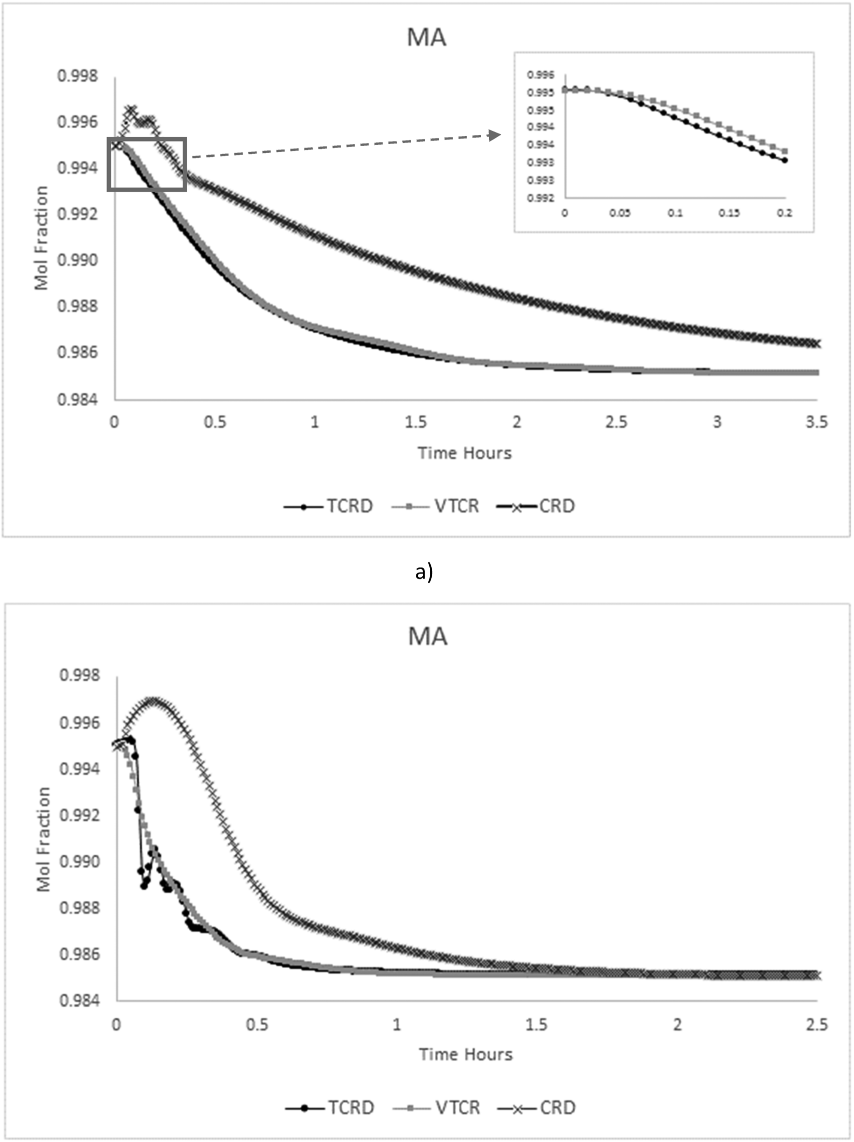
$$u_i(t) = \overline{u}_i + k_C^{ij}(y_j(t) - \overline{y}_j) + \frac{k_C^{ij}}{\tau_I^{ij}}\int_0^t (y_j(\theta) - \overline{y}_j)\,\mathrm{d}\theta \qquad (13)$$

the tuning relationships in Zavala-Guzmán et al.[18] are recalled and applied straightforwardly.

$$k_C^{ij} = \frac{2n-1}{K_{ij}}, \qquad \tau_I^{ij} = \tau_P^{ij}(\xi_{ij})^2\left(\frac{2n-1}{n^2}\right) \qquad (14)$$

As it can be observed, the relationships are given in terms of four parameters: (i) a static gain (K_{ij}), (ii) a time constant (τ_P^{ij}), (iii) a damping factor (ξ_{ij}), and (iii) a parameter of fine adjustment (n). The parameter n can be seen as the number of times it is desired for the response velocity of the closed-loop behavior to be faster than the corresponding open-loop behavior. Operatively speaking, the application of the tuning relationships is as follows (Figure 4):

1. Identify K_{ij} and τ_P^{ij} through a reaction curve of y_j yielded by a small step change in the input u_i. Particularly τ_P^{ij} is set as a quarter



a)

b)

Figure 6. (a) Dynamic responses with the same *n* value in the MA loops. (b) Dynamic responses with different *n* values in the MA loops.

of the settling time; i.e., the time elapsed from the application of an ideal instantaneous step input to the time at which the output reaches the new steady state. The smaller the change in u_i as possible to distinguish a y_j evolution, the more precise the values of K_{ij} and τ_P^{ij}, with respect to the ones that would be obtained from an analytical linearization of a mathematical model of the process.

2. Set the damping factor ξ_{ij} considering it gets the same insight as in a linear second order system; i.e., small values would yield a fast response, but oscillatory. In a linear second order system, a value of 0.8412 yields an overshoot of 5%.

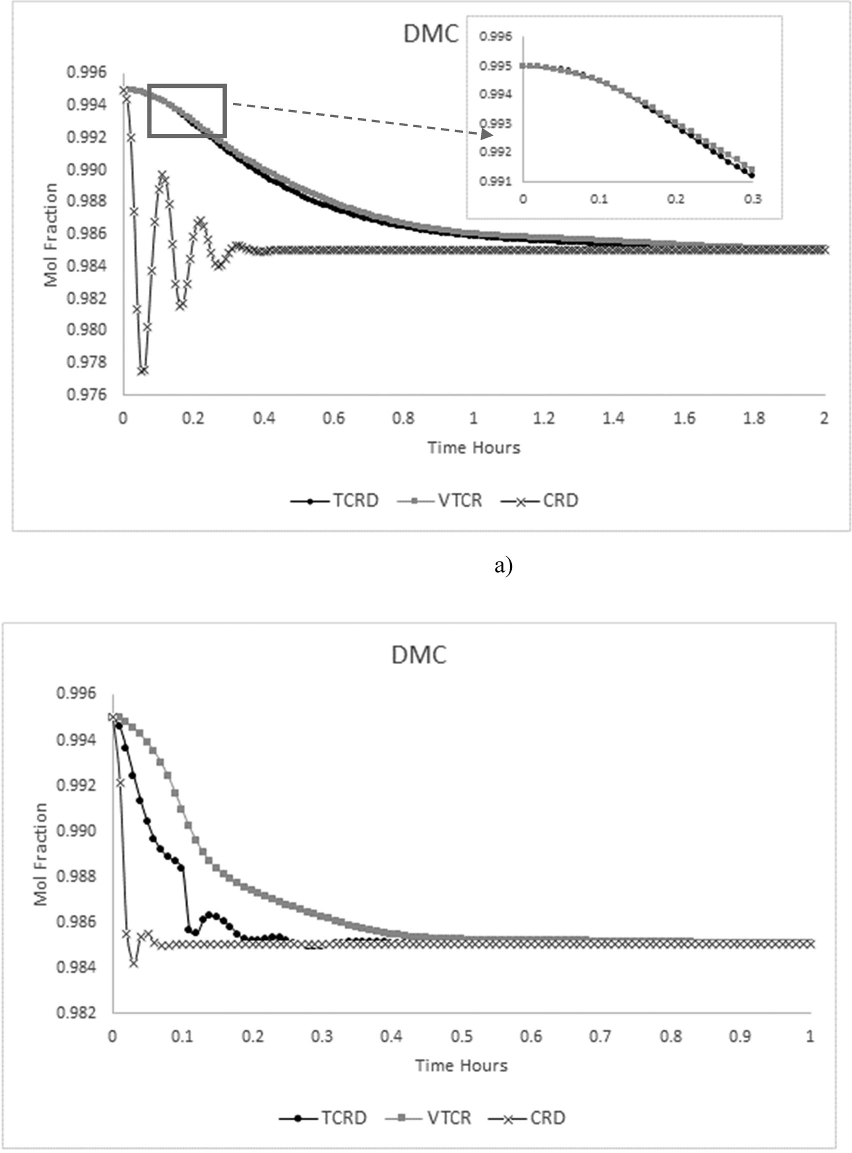
3. Tune through n. As an initial trial, set $n = 1$, calculate the controller gains, and evaluate the performance of the control system. In distillation systems, usually to evaluate the control system performance the integral absolute error (IAE) is used as the performance index in a framework of a servo-control problem.[29] Next, increase the value of n as long as the performance index diminishes.

Provided the static gain and the time constant for every control input−control output pair (step 1), steps 2 and 3 can be simultaneously applied for all the control loops, which implies that n is left as a final single tuning button for all the controllers. The performance test is initialized with $n = 1$, and additional adjustments could be done by only changing n. In this way, the tuning will be systematically carried out.

6. RESULTS AND DISCUSSION

In order to test and illustrate the effectiveness of the approach described above, the physical characteristics and process conditions for the reactive distillation systems of this study (Figures 1−3) were recalled from Contreras-Zarazúa et al.[27] Table S2 shows the nominal values of inputs and outputs; it can be noticed that the products are of high purity. Measurement delay might be a significant factor, since this system involves mass and energy transportation. However, as Luyben and Yu[5] indicate, this factor can be omitted in preliminary control studies.

In the step of the establishment of the control configuration, the relative gains and the operability indexes were computed; these outcomes are given in Tables 1 and 2, respectively.



a)

b)

Figure 7. (a) Dynamic responses with the same *n* value in the DMC loops. (b) Dynamic responses with different *n* values in the DMC loops.

Table 4. Tuning Parameters and Controller Gains for Different *n* Values

	CRD					
	k_P	τ_P (h)	ξ	n	k_C	τ_I (h)
(RDR, DPC)	0.5772	2.3835	0.8412	50	171.5219	0.0668
(RDC, DMC)	2.1724	9.3722		30	27.1585	0.4348
(RRC, MA)	0.0770	4.2352		10	246.6597	0.5694
	TCRD					
	k_P	τ_P (h)	ξ	n	k_C	τ_I (h)
(RDR, DPC)	4.5136	1.7093	0.8412	100	44.0894	0.02407
(RDC, DMC)	0.2243	2.6962		30	263.0717	0.12507
(RRC, MA)	0.2906	4.2987		30	203.0395	0.19941
	VTCR					
	k_P	τ_P (h)	ξ	n	k_C	τ_I (h)
(RDR, DPC)	4.4598	1.4421	0.8412	20	8.4533	0.0911
(WDC, DMC)	0.2134	2.9703		30	267.0781	0.1456
(RRC, MA)	0.3788	4.6734		30	187.7654	0.2122

By choosing the pairs with relative gains close to 1 and with the lower operability index, for every reactive distillation configuration, the resulting control configuration is (RDR, DPC), (RDC−WDC, DMC), and (RRC, MA). Thus, by

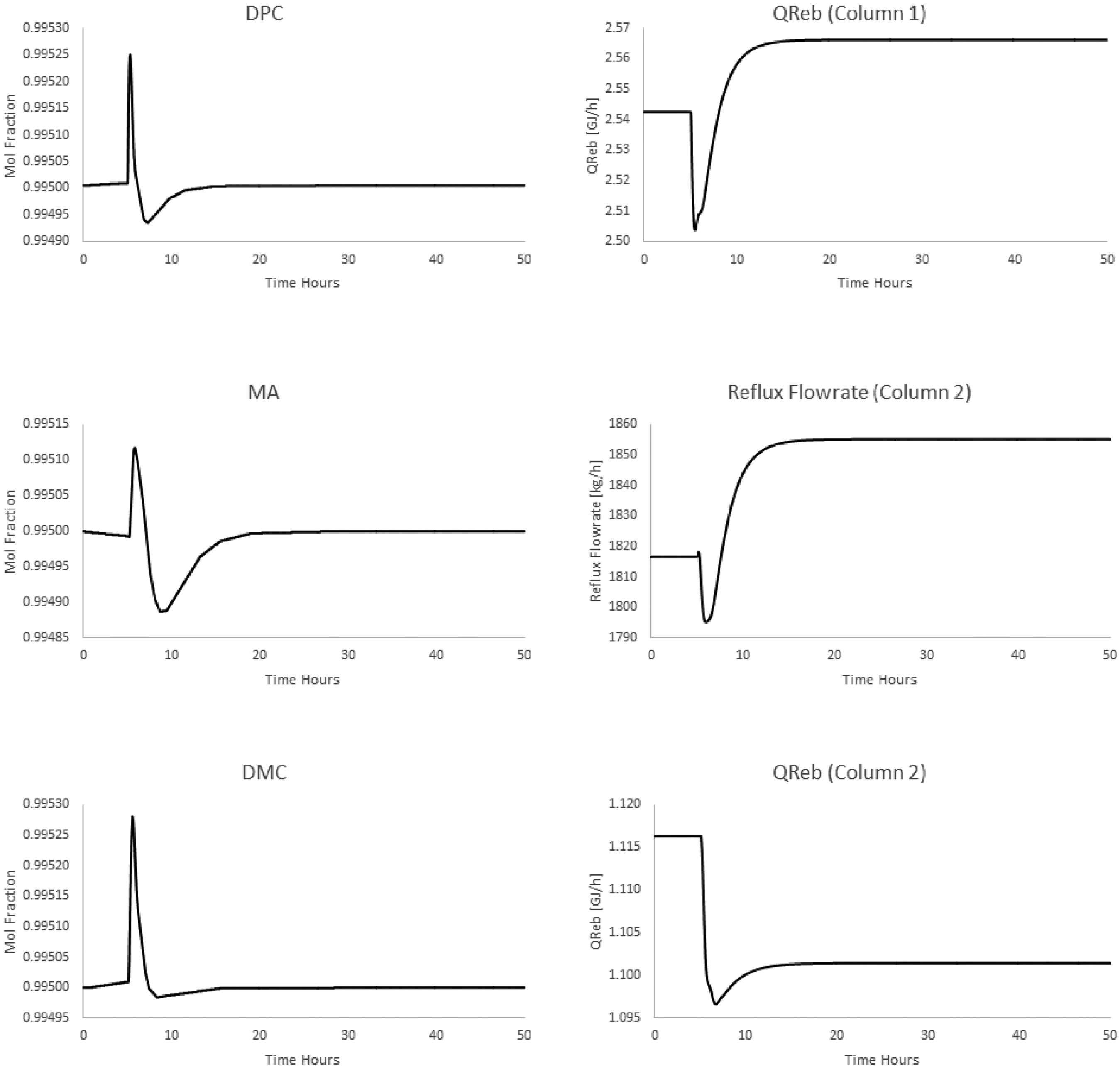


Figure 8. Responses when introducing impurity to the conventional reactive distillation (CRD) configuration.

looking at Table 1, it can be noticed that the pairing can easily be derived because several outputs only get one positive relative gain, and the input−output pairs with positive relative gains close to 1 do not overlap each other. But looking at Table 2, the OIs for CRD point out that the likely required changes in control inputs are feasible: driving a change of 1% in MA will take at least a change of 5.32% in RRC, for DMC, 4.42% in RDC, but only 0.40% in RDR for DPC. For both TCDR and VTCR, the OIs for (RDR, DPC) and (RRC, MA) pairs are even smaller than the ones for CDR; however, the OI for (RDC−WDC, DMC) pair is around 6.5 times the one for CRD, meaning that a considerable change in RDC will be taken to drive a 1% of change in DMC. Performing a change around 30% in RDC is feasible, but it seems severe. It is worth taking into account that a high purity of products is desired; thus, dealing with a 1% variation in control outputs is severe.

The resulting control loops correspond to the traditional $L-V$ arrangement in each sequence, in which the reflux flow rate L and the vapor boil-up rate V (affected directly by the heat duty supplied to the reboiler) are used to control the distillate and bottom compositions.[30]

In order to verify the feasibility of controlling every reactive distillation configuration, a scenario of the servo-control problem was considered in which a change of −1% is requested to every control output. It is worth recalling that the reactive distillations are of high purity in such a way a small change implies a considerable challenge; e.g., a positive change is not considered because it implies composition in products greater than 100%.

On the application of a tuning procedure based on relationships (eq 6), Table 3 gives the tuning parameters and resulting gains with the same damping factor ($\xi = 0.84$), and the same convergence rate button ($n = 10$) for all the controllers of every

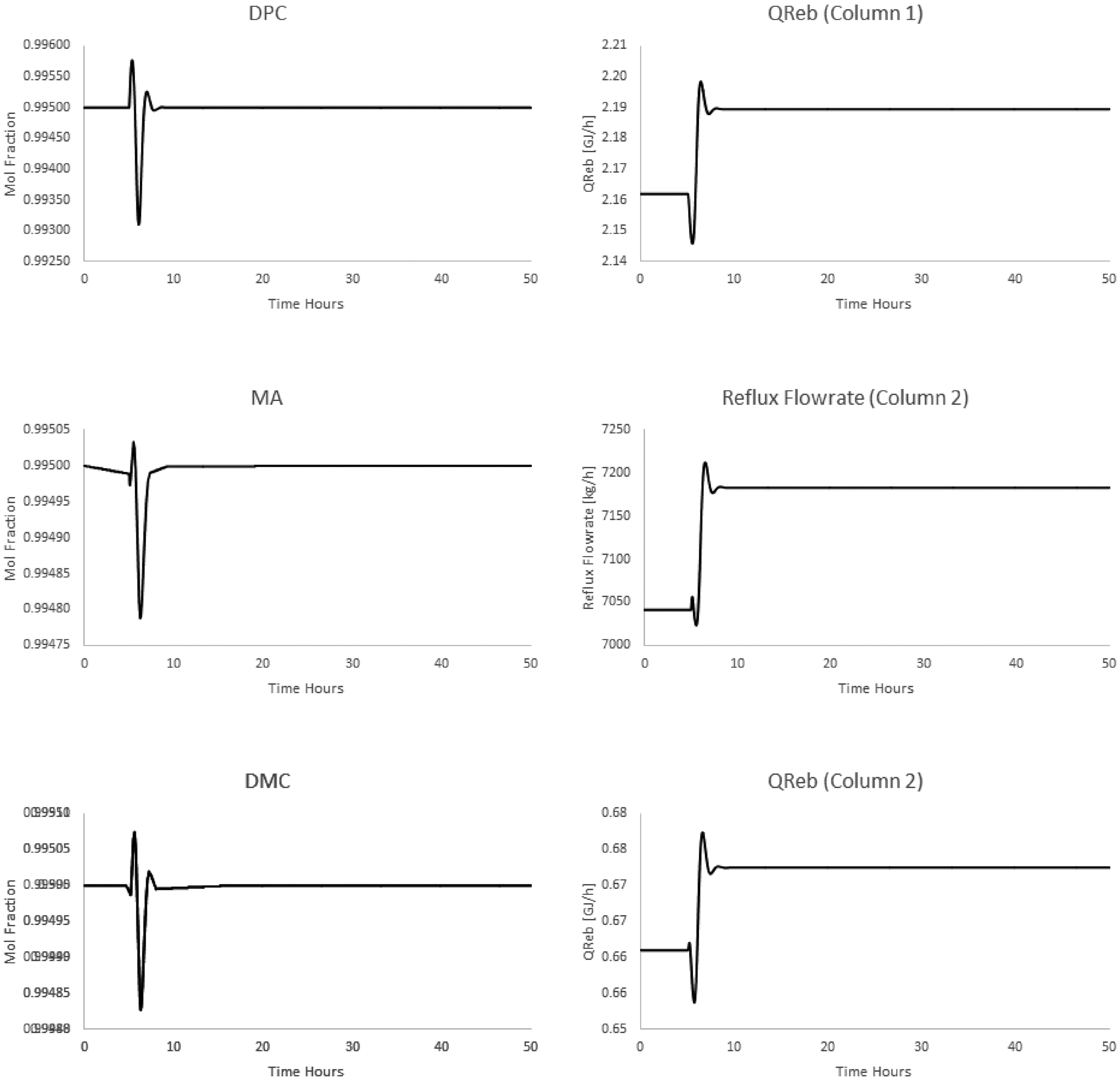


Figure 9. Responses when introducing impurity to the thermally coupled reactive distillation (TCRD) configuration.

reactive distillation configuration. In every reactive distillation configuration, the gains tuning started with $n = 1$, and the simultaneous working of all controllers yielded a smooth but slow convergence in all the control outputs. Then, in order to reduce the settling time, the n value was increased; in turn, the convergence performance was evaluated through the IAE. Figures 5a, 6a, and 7a illustrate the trajectories for DPC, DMC, and MA, respectively, in every reactive distillation configuration, and corresponding settling times, and IAEs are given in Table S3 (Supporting Information). In this framework of similar tuning parameters, controlling DPC in the simpler configuration CRD seems smoother and faster; although the convergence of DPC in the more complex configurations is oscillating, they do not present unfeasible or hasty changes. Conversely, controlling MA in CRD is slower than in the other more complex configurations, but for the three configurations the convergence is monotonic; it is noteworthy that MA in CRD exhibits inverse response, and the corresponding gains provided a convergent trajectory. With respect to DMC, the trajectories of CRD and TCRD are almost equal, and the one of VTCR exhibits oscillations, but it is the fastest one. As it can be observed, comparing convergence rates of output variables, in all the reactive distillation configurations, DMC exhibited the faster convergence and MA the slower one. In the other hand, estimating the outputs' settling times in an open-loop mode, through the constant times given in Table 3 multiplied by 4, it can be seen that the convergence times are slower.

A fine-tuning was explored by increasing n independently for each control loop, taking as initial values the ones of the previous tuning task. Table 4 gives the tuning parameters and resulting gains with the same damping factor, and a different convergence rate button for all the controllers of every reactive distillation

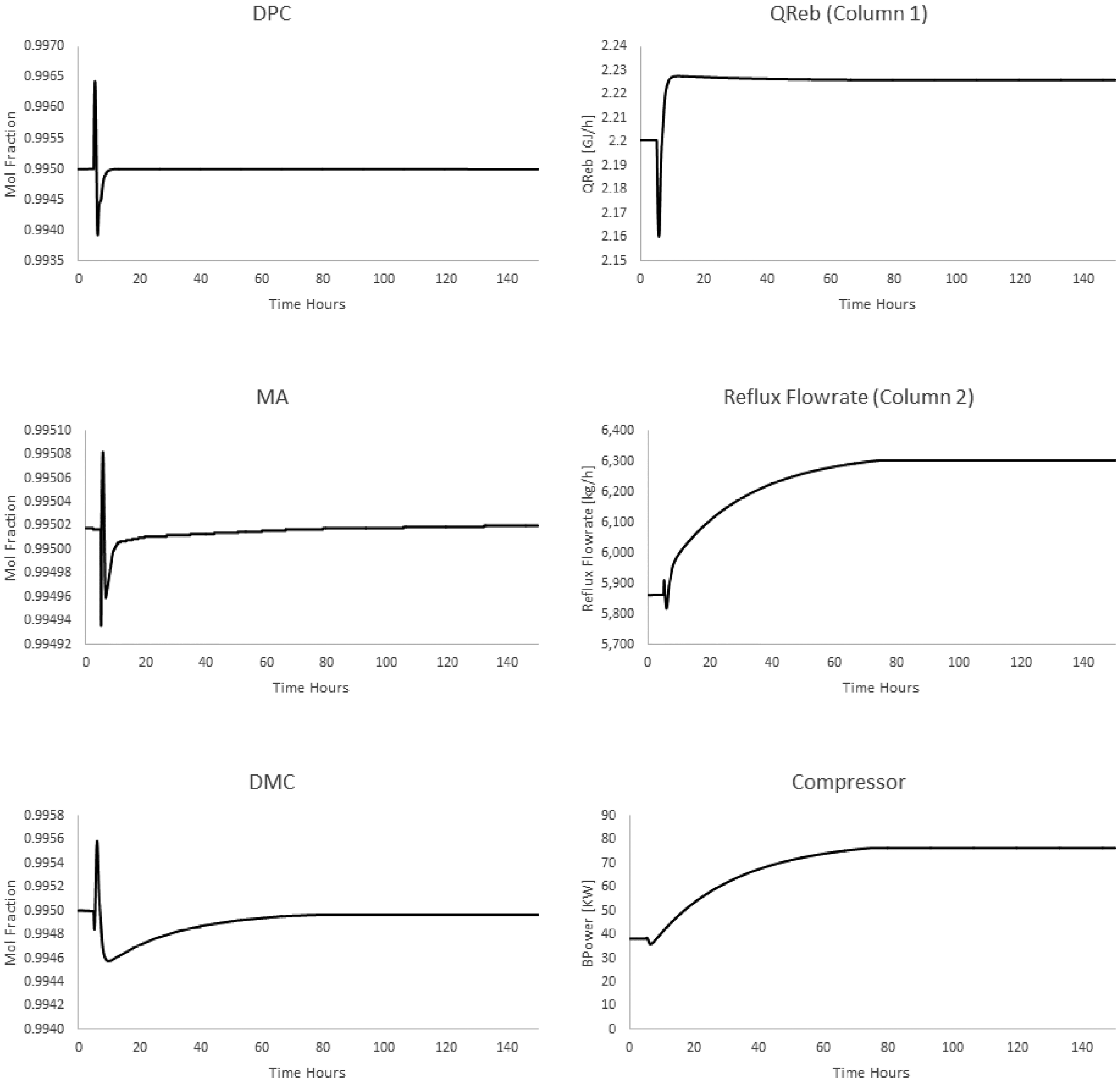


Figure 10. Responses when introducing impurity to the vapor recompression and thermally coupled reactive distillation (VTCR) configuration.

configuration. Figures 5b, 6b, and 7b illustrate the trajectories for DPC, DMC, and MA, respectively, in every reactive distillation configuration, and corresponding convergence times and IAEs are given in Table S4 (Supporting Information). Much faster convergence was achieved for each control loop in every reactive distillation configuration, and although some trajectories exhibit oscillations, their amplitudes are small. Even the performance improvement is reflected in the corresponding IAEs.

Finally, to test the control systems on rejecting disturbances, a composition control problem was performed as follows: A 0.1% impurity (DMC) was introduced into the composition of the PA feed stream, which has a direct effect on the composition of product streams. Fluctuations in the feed composition represent the most substantial upsets with which a distillation control system must deal on a continuous basis. A feed composition change shifts the composition profile through the column resulting in a considerable upset in the product compositions; in turn it can be said that distillation systems are sensitive to this kind of disturbance. Figures 8–10 show the dynamic responses in each of the study configurations. For Figure 8, we observe the behavior of each product (DMC, DPC, and MA), and their respective manipulated variables. It is visualized that every response stabilizes gradually, either in the compositions, as in the reboiler duty and the reflux ratio. The same happens in Figures 9 and 10, which shows that the three systems support changes in the feed and are able to stabilize the outputs of the products.

7. CONCLUSION

A class of reactive integrated distillation systems to produce diphenyl carbonate was feasible to be controlled, where an effective convergent behavior was provided by conventional

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linear PI controllers. The novelty of this work lay in the establishment of control configurations for complex reactive distillation systems, which are highly sensitive and with high purity products, with simple and systematic techniques: the RGA posed the control loops, and it was complemented by the proposal of an operability index to verify the practical feasibility of the control loops. Moreover, the tuning of controllers could be achieved through simple relationships coming from a stable pole assignment approach, providing the feasibility of adjusting convergent performance in a systematic and insightful way.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.iecr.9b02678.

> Kinetic parameters for reaction rate coefficients; design variables of reactive distillation configurations to synthesize DPC; settling times and IAEs of controlled systems for equal and different values of *n* (PDF)

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Notes

The authors declare no competing financial interest.

■ NOMENCLATURE

E_a = activation energy
k_0 = pre-exponential factor
τ_I = integral time
λ_{ij} = relative gain
C_j = concentration of component j (kmol/m^3)
r_i = reaction rate of the ith reaction (kmol/m^{3}s)
ξ_{ij} = damping factor
τ_P^{ij} = time constant
CRD = conventional reactive distillation
DMC = dimethyl carbonate
DPC = diphenyl carbonate
DWC = dividing-wall distillation column
IAE = integral absolute error
K_{ij} = steady-state gain
k_P = proportional gain
L = reflux flow rate
MA = methyl acetate
MIMO = multiple-input multiple-output
n = parameter of fine/final adjustment
OI = operability index
PA = phenyl acetate
PI = proportional integral
PID = proportional−integral−derivative
QVR = heat supplied by the heat exchanger
RDC = reactive distillation column
RDC = reboiler duty of the recovery column
RDR = reboiler duty of the reactive column
RGA = relative gain array
RRC = reflux ratio of the recovery column
RRC = reflux ratio of the recovery column
R−V = reflux/boil up
SVD = singular values decomposition
TCRD = thermally coupled reactive distillation
u_i = control inputs
V = vapor boil-up rate
VTCR = vapor recompression and thermally coupled reactive distillation
WDC = work duty in the compressor
y_i = control outputs

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