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Published in:
AIChE Journal

Link to article, DOI:
[10.1002/aic.17227](https://doi.org/10.1002/aic.17227)

Publication date:
2021

Document Version
Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):
Iftakher, A., Mansouri, S. S., Nahid, A., Tula, A. K., Choudhury, M. A. A. S., Lee, J. H., & Gani, R. (2021). Integrated Design and Control of Reactive Distillation Processes Using the Driving Force Approach. *AIChE Journal*, 67(6), [e17227]. <https://doi.org/10.1002/aic.17227>

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Integrated Design and Control of Reactive Distillation Processes Using the Driving Force

Approach

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Abstract Superior controllability of reactive distillation (RD) systems, designed at the maximum driving force (design-control solution) is demonstrated in this paper. Binary or multi-element single or double feed RD systems are considered. Reactive phase equilibrium data, needed for driving force analysis and design of the RD system, is generated through an in-house property prediction tool. Rigorous steady-state simulation is carried out in ASPEN plus in order to verify that the predefined design targets and dynamics are met. A multi-objective performance function is employed to evaluate the performance of the RD system in terms of energy consumption, sustainability metrics (total CO₂ footprint), and control performance. Controllability of the designed system is evaluated using indices like the relative gain array (RGA) and Niederlinski

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process which may lead to differences between this version and the [Version of Record](#). Please cite this article as doi: [10.1002/aic.17227](https://doi.org/10.1002/aic.17227) © 2021 American Institute of Chemical Engineers
Received: Sep 07, 2020; Revised: Dec 08, 2020; Accepted: Jan 10, 2021

Index (N_I), to evaluate the degree of loop interaction, as well as through dynamic simulations using proportional-integral (PI) controllers and model predictive controllers (MPC). The design-control of the RD systems corresponding to other alternative designs that do not take advantage of the maximum driving force is also investigated. The analysis shows that the RD designs at the maximum driving force exhibit enhanced controllability and lower carbon footprint than the alternative RD designs.

Keywords Reactive Distillation, Model Predictive Controller, Driving Force, Integrated Design-Control, Process Systems Engineering

1. Introduction

Reactive distillation (RD) takes advantage of the synergy created when combining heterogeneous/homogenous catalytic reactions or non-catalytic reactions and separation into a single unit¹. RD improves the productivity and selectivity by primarily simultaneous removal of products from reactants as well as suppressing side reactions², reduces capital cost as well as the need for solvents³, avoids/degrades azeotropes⁴, reduces energy usage by using the heat of exothermic reaction in-situ for the vaporization of the liquid, resulting in lower CO₂ emission and less waste⁵, among others. These advantages not only attract industrial perspectives and applications⁶ but also make RD a suitable intensified process to combat ever-increasing worldwide energy demand, which is expected to increase about 3.5 times (7 times increase in electricity demand) to sustain the ever-increasing population⁷. The potential of RD technology, however, has not yet been fully tapped, and there is still ongoing research to improve it further by other means, for example, ultrasound or microwave-assisted RD⁸, use of high-gravity fields (HiGee)⁹, and/or addressing issues such as operability and control¹⁰. There have been more than 150 successful industrial applications of RD technology¹¹, for example, for selective

hydrogenation of mixed hydrocarbons, selective desulfurization of mid catalytic naphtha, and isomerization of n-olefins to iso-olefins. In particular, RD systems have been reported for production of methyl acetate⁴, production of fuel additives or oxygenates (MTBE, ETBE, TAME)⁶; toluene disproportionation to benzene and o-xylene¹², hydrolysis of methyl acetate¹³, biodiesel production¹⁴, production of ethylene glycol¹⁵, production of silanes¹⁶, and many more.

Substantial amount of research has been done on the synthesis and/or design methods for RD systems over the years¹. These include conceptual synthesis and design^{17,18}, phenomena-based method approaches¹⁹, optimal design using deterministic and stochastic techniques²⁰ and superstructure optimization²¹, graphical methods¹², membrane-assisted reactive distillation processes²², boundary value methods²³⁻²⁵, pinch based methods²⁶, shortest stripping line method²⁷, to name a few.

Several control studies involving RD operations have been reported¹. For the control of ethyl acetate production, Chien et al.²⁸ employed a PI(D) controller to control tray temperature, while Vora and Daoutidis²⁹ applied linear and nonlinear control strategies. For the control of ethylene glycol production, Heath et al.³⁰ employed a PI controller while Kumar and Daoutidis³¹ considered output multiplicity and applied nonlinear inversion based controller. Wang et al.³² controlled tray temperature using a PI controller for diethyl carbonate synthesis. Al-Arfaj and Luyben³³ studied the use of a PI controller to control tray temperature and product purity for methyl acetate production. Medina-Herrera et al.³⁴ worked on trichlorosilane decomposition and employed temperature control to ensure on-spec production of multiple products.

Applications of advanced control algorithms such as model predictive controller (MPC) are also reported for RD operation in the literature. Lee³⁵ has reviewed the development and application of MPC. Some of the notable works include: controlling product purity in a TAME synthesis

process³⁶; applying nonlinear MPC with a neural network model for controlling product purity of a methyl acetate RD system³⁷. In the case of ethyl acetate production, Kawathekar and Riggs³⁸ applied nonlinear MPC for controlling product purity; Venkateswarlu and Reddy³⁹ applied nonlinear MPC based on stochastic optimization for controlling product purity, reboiler & condenser holdups, and column pressure; Balasubramhanya and Doyle⁴⁰ applied nonlinear MPC to control tray temperature.

It is well-known that process design and process control, when considered independently, generally lead to conflicting objectives for the chemical processes, e.g., over design versus not robust design, or, infeasible versus economically suboptimal operating points⁴¹. Therefore, design and control issues are best addressed simultaneously (as in integrated process design and control) to obtain economically attractive, yet operationally feasible, and flexible processes. One of the convenient features of the simultaneous approach is the possibility to identify and eliminate potentially promising design alternatives that may have controllability problems due to a loss of degree of freedom, which shrinks the controllability region⁴². Various aspects of the simultaneous approach have been studied⁴³⁻⁴⁵. To name a few, simultaneous design and control of acidic acid production employing a PI(D) controller has been reported by Georgiadis et al.⁴⁶ and Pistikopoulos and Sakizlis⁴⁷. For ETBE production, Sneesby et al.⁴⁸ explored the interactions between design and control, while Bernal et al.⁴⁹ considered phenomena such as column hydrodynamics and pressure drop across the column together with a rigorous dynamic model for simultaneous design and control. Chung et al.⁵⁰ studied the design and control of a reactive distillation process for esterification of levulinic acid and n-butanol. However, work on integrated design-control using MPC has scarcely been reported. Francisco et al.⁵¹ reported a methodology for the simultaneous design of processes and linear MPCs, providing the plant

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dimensions, the control system parameters, and a steady-state working point simultaneously; Bahakim and Ricardez-sandoval⁵² proposed a methodology based on stochastic simultaneous design and MPC control of chemical processes under uncertainty. Mansouri et.al⁵³ have shown that by simultaneously considering design and control issues through the driving force concept^{12,54}, optimal design-control of reactive distillation units can be achieved. Here the driving force is defined as the difference in composition of a specific element between two coexisting phases and the plot of driving force versus liquid phase composition of component in a non-azeotropic binary mixture has a well-defined maximum. The main takeaway from this work is that a distillation column, whose feasibility of operation depends on the driving force, when designed on the basis of the maximum available driving force, would consume the least energy and also would be the easiest to operate^{54,55}. Recently, Lopez-Arenas et al.⁵⁶ proved theoretically that an RD column designed for operation at the maximum available driving force indeed corresponds to the minimum energy requirement. This means that if the maximum driving force can apriori be identified, the corresponding minimum energy consumption and easy operation can be obtained without formulating and solving an optimization problem for the design and therefore control of an RD column.

In this work, the development and application of a versatile framework for systematic integration of the driving force concept for design with various control methods, such as PI(D) and MPC, are presented. The results reported by Mansouri et al.⁵⁷ are confirmed and extended through this framework. The main novelties of this work are as follows: 1) the extended framework now covers additional design-control options so that new RD systems in terms of configuration as well as reaction systems can be more easily introduced; 2) reactions are now not limited to only the liquid phase; 3) application and evaluation of advanced controllers such as MPC is now

possible; 4) the extended framework has been converted to a computer-aided toolbox that guides the user through the steps of the work-flow of the integrated design-control method, provides access to in-house and/or external tools needed at each step, and, transfers the data from one step to another.

The extended framework allows for different configurations of the RD column and integrates the controller designs through a common set of variables that define both the design and control of the RD column operation. For example, product purity (a specification for process design also serves as a set-point value for the controller) is paired with reflux ratio (which is optimized in process design also serves as a reference value for the manipulated variable) at the maximum achievable driving force. For a reactive system, the toolbox based on the framework guides the user through the steps of the design and control algorithms and sets up the closed-loop simulation for verification. Application results for six RD column operations reported previously by others are presented together with the advantages of using the framework and toolbox.

This paper is organized as follows: First, an overview of the integrated design-control framework is presented. Next, the details of six case studies illustrating the application of the framework and toolbox are presented. Next, key results are discussed and the detailed design and closed-loop dynamics are presented for one case in the main manuscript, while the detailed results for the other case studies along with algorithms and mathematical derivations are given as supporting material. **2. Integrated Process Design-Control Framework**

The framework for driving force based integrated design and control of RD processes is illustrated in Figure 1. This framework consists of three main steps: (1) Data collection and problem formulation; (2) Integrated design-control; (3) Verification. The integration of process design and control objectives is achieved in step 2 through operation at the maximum available

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driving force, which is achieved by identifying variables that can be assigned dual roles (a process design role and a controller design role) and whose values are determined from a driving force diagram of the reacting system. Table 1 lists a selection of these variables. The verification in step 3 is carried out through simulations in open-loop and closed-loop to evaluate the performance of the designed reactive distillation process with and without the controllers. This framework has been implemented in a new version of ProCADC⁵⁸, an integrated software toolbox for process design and control, which also provides a link to needed property estimation tools as well as links to external tools (ASPEN PLUS, MATLAB). All case study results presented in this paper are obtained using this toolbox.

[Figure 1]

[Table 1]

Step 1: Data collection and Problem formulation

Step-1a: RD problem data and/or specification: Data/information on raw materials, products, catalysts, reaction conversions, and feed conditions (temperature, pressure, and composition) are collected, and design targets and product specifications are given. An objective function, which could be maximized (or minimized) from both the design and control perspectives, is defined. Note, however, with the driving force concept, a numerical optimization solver is not employed. Instead, the objective function serves to evaluate a multi-variable performance function at different design-control scenarios as the minimum in terms of energy consumption and ease of operation are related to the apriori located maximum driving force of the reacting system.

Step 1b: (Reactive system representation): In this step, the number and identity of elements needed

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to represent the reactive system are determined through the algorithm of Pérez-Cisneros et al.⁵⁹. A revised version of this algorithm is given in section 2 of Supplementary Material as Algorithm-1s. The elements are transformed variables, proposed first by Ung and Doherty⁶⁰ to reduce the dimensionality of the reactive system and thereby ease of visualization of the phase behavior. Later, Pérez-Cisneros et al.⁵⁹ proposed algorithms for the simultaneous calculation of chemical and physical equilibrium by formulating the Gibbs energy minimization problem in terms of elements, where elements are atoms or groups of atoms and identified by the minimum set needed to represent the reacting system and to satisfy the atomic balance for all compounds (including inert compounds). Usually, it is the number of compounds (reactants and products) minus the number of reactions plus the number of inert compounds. If a binary element system is encountered, go to step-2; otherwise, go to *Step 1c* to identify the key pair of elements for the multi-element system.

Step 1c: (Identify the key elements): The equivalent binary elements, that is the light key and heavy key elements, are selected according to the rules of key element selection for a multi-element system⁶¹, which are given in section 1 of Supplementary Material as Rules set-A.

Step 2: Integrated design-control

The objective of this step is to determine the RD process design¹² (*Step 2a*) at the maximum driving force and the corresponding controller design.

Step 2a (Design issues) - Step 2a.1 (Generate reactive vapor-liquid equilibrium data): The reactive vapor-liquid equilibrium (VLE) data is either obtained from experiments or computation of reactive bubble points and/or dew points, for which the algorithm of Daza et al.,¹² which is available through the new toolbox in ProCADC⁵⁸, is employed. This implemented

algorithm in the toolbox generates binary and equivalent binary element reactive phase diagrams and is included as Algorithm 2s in section 2 of Supplementary Material. Note that in this algorithm, the choice of discretization may affect the accuracy of the results. Since the calculation of the reactive VLE data is not computationally expensive, one can decrease the discretization interval (in absence of miscibility gaps) to obtain a smoother curve.

Step 2a (Design issues) - Step 2a.2 (Reactive distillation design based on binary or equivalent binary elements): The objective of this step is to determine the RD column design (i.e., number of stages, reflux ratio, and feed location) based on binary or equivalent binary element at the maximum driving force. The steps required to perform this task are given in Algorithm 3s (section 2 of Supplementary material) for a single feed RD column; and Algorithm 1 (described in detail in section 2.1) for a double feed RD column. The last step of Algorithm 3s or Algorithm 1, involves steady-state simulation of the designed process to verify that the design objectives are satisfied.

The reactive driving-force based on the binary elements (or binary equivalent elements) is calculated using Equation 1 as described by Daza et al.¹², or in the case of equivalent binary elements, light key equivalent element compositions are used to calculate the driving force.

$$DF = W_i^v - W_i^l = \frac{W_i^l \alpha_{ij}}{1 + W_i^l (\alpha_{ij} - 1)} - W_i^l \quad (1)$$

As part of Algorithm 3s (section 2 of Supplementary Material), the reactive McCabe-Thiele algorithm is given as a sub-algorithm, which is applied to calculate the minimum number of stages (corresponding to the maximum available driving force) to obtain the desired product specifications (targets) defined in step 1. The calculation is based on the well-known McCabe and Thiele method⁶² for non-reactive distillation design. Daza et al.¹² extended this method to

include reactive binary element distillation columns, which can also be used for equivalent binary element columns.

For a double feed RD column with feed flowrates H and K , Algorithm 1 is applied to design the column at the maximum driving force. Note that this algorithm is adapted from the original McCabe-Thiele method⁶² for the design of distillation columns with two feeds.

Step 2b (Optimal design-control issues): In this step, the control structure is selected with respect to two main criteria (sensitivity of controlled variables with respect to disturbances in the feed and sensitivity of manipulated variables to controlled variables) that are highlighted in Algorithm 4s (section 2 of Supplementary Material). The performance of the control structure is verified at the maximum driving force analytically using Algorithm 4s (section 2 of Supplementary Material). The application of this concept is analytically illustrated by Mansouri et al.,^{53,57} where it is shown that the sensitivity of controlled variables with respect to feed disturbance is minimum, and the sensitivity of controlled variables with respect to manipulated variables is maximum for RD columns designed at the maximum available driving force. These two conditions ensure minimum control moves. Thus, the term ‘optimal’ in this work is used in a sense that the summation of control moves required is smaller than at other design (see results in section 4.2).

Step 3: Verification through simulation and analysis

The objective of this step is to verify the performance of the integrated design-control at the maximum driving force of the RD system through analysis of steady-state and dynamic simulation results. Figure 2 shows the process design – controller block diagram employed for verification steps.

[Figure 2]

Step 3a - Control structure verification: The applicability of the control structure obtained at the maximum binary element or equivalent binary element driving force, is verified by applying Algorithm 5s (section 2 of Supplementary Material). The control structure is verified by inspecting independently, the Relative Gain Array (RGA) values and the Niederlinski Index (N_I) values. The stability of the RD systems is checked by the eigenvalue analysis of the open-loop and closed-loop dynamic models.

Step 3b - Dynamic closed-loop analysis: The objective of this step is to evaluate the closed-loop performance of the control structure identified and verified in *Step 2b* and *Step 3a*, respectively, in the presence of disturbances by applying Algorithm 6s (section 2 of Supplementary Material). The disturbance is introduced through feed flow rate (and composition).

Step 3c – Selection: In this step, the value of each term of the performance (objective) function (see Eq. 2) is calculated.

2.1 Algorithm 1: Driving force based RD design for a two feed column

The objective is to find the RD column design (number of stages, reflux ratio, feed location) with two feeds at the maximum driving force for the specified design targets. This algorithm is an adopted version, proposed earlier by Mansouri⁵⁵.

Step (i): Retrieve vapor-liquid equilibrium element data (for the binary or equivalent binary system using Algorithm 2s – see section 2 of Supplementary material).

Step (ii): Calculate the driving forces for the entire composition domain using Equation (1), then plot $|DF|$ versus W_i^l based on the light key element (or the equivalent light key element).

Step (iii): Identify the area of operation of the driving force diagram (reactive zone information), i.e., feed compositions (feed one and two), distillate (W^D) and bottom (W^B) compositions based on the light key element (or the equivalent light key element) using design targets set in Step 1.

In the text above, W_k and W_h are the compositions of the feeds; W_g is the composition of feed-g if the mixture with the two feeds are mixed such that it corresponds to the maximum driving force on the x -axis of the driving force diagram (D_x). Note also, if $W_h > W_k$, then the H feed is placed higher in the RD column. If the area of operation is not between 0 and 1 on the x -axis, then a re-scale of the x -axis between 0-1 is performed

Step (iv): Determine the reflux ratio and boil-up ratio (similar to *Step (iv)* of Algorithm 3s).

Step (v): Construct the PTXY diagram using the vapor-liquid equilibrium (binary or equivalent binary) element data from Algorithm 2s.

Step (vi): Draw the angle bi-section line (45° line), and locate W^D, W^B, W_h, W_k , and W_g on the x -axis of the PTXY diagram.

Step (vii): The enrichment line for the middle of the column is found by joining the points where enrichment lines for the rectifying and stripping sections intersect the lines $x = W_h$ and $x = W_k$. Note that the rectifying and stripping lines from *Step (iv)* are exactly the same

as in the case of a single feed (see Algorithm 3s), i.e., they start from the top and bottom product compositions.

Step (viii): Find the minimum number of reactive stages by drawing the steps for stages.

Step (ix): Perform a steady-state simulation. If simulation indicates that the specified product composition has not been reached, then add non-reactive stages one-at-a-time to the top and bottom of the reactive section until the desired purification of the products is achieved.

3. Application Examples – Case Study Data

The framework for integrated process design and control of single feed binary and multi-element systems, and double feed multi-element system has been tested through the following six case studies 1) production of Methyl-tert-Butyl-Ether without the presence of inert, 2) production of Methyl-tert-Butyl-Ether in the presence of inert, 3) production of methyl acetate, 4) toluene disproportionation, 5) production of Ethyl-tert-Butyl Ether and 6) production of Tert-Amyl-Methyl-Ether. It is assumed that equilibrium is achieved as the chemical reaction taking place is fast. In this section data related to step 1 of the framework is given.

3.1 Step 1: Data collection and problem formulation

Step-1a: The key information of *Step-1a* for the six cases is listed in Table 2, and the data collection for case study 3 is described in detail. Detailed description for the other cases is given in section 3 of Supplementary Material. For the production of methyl-acetate via a reactive distillation column (case study 3), two feeds having same flowrates, but one containing methanol and the other acetic acid are considered. Also, reactive and non-reactive sections are considered. The design objective in this section is to obtain the RD column design operating at the maximum driving force to produce high purity methyl-acetate. The reaction between

methanol (MeOH) and acetic acid (HOAc) yields methyl acetate (MeAc) and water (H₂O). The reaction takes place in the liquid phase over a catalyst⁶¹. It is exothermic with heat of reaction of −5.42 kJ/mol and is given as follows:

The design targets for all the case studies are given in Table 1s-6s (section 4 of Supplementary Material). For case study 3, the design target for the reactive section of the RD column is specified - data is obtained from Jantharasuk et al.⁶¹. In order to verify the integrated design and control, a multi-objective performance function involving a set of metrics to evaluate control performance is considered, as given below:

$$f_{obj} = \text{minimize } J_1, J_2, J_3, J_4, J_5, J_6 \quad (2)$$

where, J_1 = energy consumption associated with the process, J_2 = integral absolute error (*IAE*), J_3 = total variation of the manipulated variables, J_4 = relative gain array (RGA), J_5 = Niederlinski Index (N_I), and J_6 = total CO₂ footprint per kg feed.

Note that this is a multi-objective performance function, which takes into account both design objectives and control objectives. The values of steady-state metrics, J_1 and J_6 , measure energy-efficiency and environment friendliness of the process. J_2 and J_3 are controller performance metrics which are defined below:

$$J_2 = IAE = \int_0^{\infty} |y - y_{sp}| dt \quad (3)$$

$$J_3 = TV = \sum_{i=1}^{\infty} |u_{i+1} - u_i| \quad (4)$$

The diagonal values of J_4 indicate the degree of interaction between control loops. J_5 indicates the system stability. Note that for a 2×2 system, N_I and RGA are clearly related because $N_I = 1/(1,1)_{RGA}$. Hence, the condition is that both have to be positive. Both metrics are included here to check the appropriateness of the control structure. The aim is not to get a weighted scalar value of this function but to evaluate each of the metrics separately and verify that J_1, J_2, J_3 and J_6 are the lowest, while J_4 and J_5 satisfy the controllability criteria for a design at the maximum driving force.

Step 1b - Binary element reactive system representation: For case studies 1, 4, 5 & 6, the number of compounds is three and the number of reactions is one. So, the reactive system can be represented by two elements. The element matrix, choice of elements, and element reaction are given in Table 7s (section 5 of Supplementary Material).

Step 1c - Identify the key elements: For case studies 2 & 3, the number of compounds is four and the number of reactions is one. So, these reactive systems can be represented by three elements. The element matrix, choice of elements, and element reaction for case studies 2 and 3 are given in Table 8s (section 5 of Supplementary Material) and Table 3, respectively. The key elements are chosen by following the Rules Set-A (section 1 of Supplementary Material). For both case studies, element C is selected as the non-key element. For case study 2, B and A are selected to be the light key (LK) and the heavy key (HK), respectively. For case study 3, element A is the light key element (LK) and element B is the heavy key element (HK).

[Table 2]

[Table 3]

4. Applications – Case Study Results and Discussion

In this section, the results from the application of the framework and the toolbox are given in detail for case study 3 together with some highlights related to the other case studies. Detailed results for the case studies are given in Supporting Material (see sections 6-8).

4.1 Step 2: Integrated design-control at maximum driving force (IDC-MDF)

Step 2a - Design issues (Step 2a.1: Generate reactive vapor-liquid equilibrium data): The reactive VLE data for the six case studies are generated through Algorithm 2s by selecting the appropriate thermodynamic models. The data used for the six case studies are given in Table 9s (section 6 of Supplementary Material). A discretization interval of 0.05 is used in this work to calculate the VLE data. The calculated reactive bubble point for the entire composition space for all the six case studies are given in Figure 1s (section 6 of Supplementary Material). These represent the $T - W_A^l - W_A^v$ phase diagrams for binary element reactive systems or the $T - W_{LK,eq}^l - W_{LK,eq}^v$ phase diagrams for multi-element reactive systems.

Step 2a - Design issues (Step 2a.2 - Reactive distillation design based on binary or equivalent binary elements): The generated reactive VLE data in terms of compounds are converted to reactive driving force diagrams, as illustrated in Figure 3. Subsequently, the RD columns are designed at the maximum binary or equivalent binary element driving force. Note that, in the reactive driving force diagram (see Figure 3), point A denotes the location of the driving force for the column design at the maximum driving force, whereas point B denotes the location of the driving force for alternative design 1 (of which the design parameters are given in Table 8). The area of operation is identified on the x -axis of the reactive driving force diagram in terms of the light key element (for a binary element system) or equivalent light key element (for a multi-element system).

[Figure 3]

Operating area is defined by selecting light key or equivalent light key element distillate composition (W_A^D or $W_{LK,eq}^D$) and light key or equivalent light key element bottom composition (W_A^B or $W_{LK,eq}^B$) by following Algorithms 3s (section 2 of Supplementary Material) for single feed binary element systems. The points D_x and D_y corresponding to the maximum driving force are identified and consequently, the slopes of these operating lines are determined and correspond to the minimum reflux ratio (RR_{min}) and reboil ratio (RB_{min}). Therefore, the actual RR and RB values are obtained by multiplying RR_{min} and RB_{min} by 1.2, giving the new slopes of the operating lines. The number of stages (N) is calculated for all case studies by applying the reactive McCabe-Thiele method. The results of the application of the reactive McCabe-Thiele method for all the case studies are given in Figure 2s-7s (section 7 in the Supplementary Material).

For case study 3, since it is a two feed and multi-element system, the area of operation is identified on the x -axis of the reactive driving force diagram in terms of the equivalent light key element. The feed and design target compositions, which are converted to element basis are translated to equivalent element compositions. Note that the area of operation is rescaled between 0-1 in the composition domain on the x -axis of the driving force diagram according to Algorithm 1. Note that, the composition of the feeds are W_h and W_k ; and the design targets are the distillate and bottom compositions (W^D, W^B). The light key equivalent element is in pure state in one feed ($W_k = 1$) and does not exist in the other feed ($W_h = 0$). The feed (with methanol) that contains the light key element is introduced at the last reactive stage and the other feed (with acetic acid) that does not contain the light key element is introduced at the first

reactive stage (counting from the top) to ensure countercurrent flow. Non-reactive stages are added to the top and bottom of the reactive zone, one at a time until the design targets are satisfied. The total number of stages includes the number of the reactive stages and non-reactive stages. Note that the reboiler and condenser are considered as non-reactive stages. Table 4 presents the RD column design parameters at the maximum driving force for the six case studies.

[Table 4]

Step 2b: Optimal design-control issues

The controlled variables ($\underline{y}$) are the top and bottom compositions, and the manipulated variables ($\underline{u}$) are the reflux rate and the reboiler duty, which for all the case studies are listed in Table 10s (section 7 in the Supplementary Material). The values of dDF/dW_A^l (for the binary elements) or $dDF_{LK,eq}/dW_{LK,eq}^l$ (for the multi-elements) are calculated and plotted versus W_A^l (binary element)

or $W_{LK,eq}^l$ (multi-element) for the six case studies as shown in Figure 8s (section 7 in the Supplementary Material). Note that the values for dDF/dW_A^l (for the binary elements) or $dDF_{LK,eq}/dW_{LK,eq}^l$ (for the multi-elements) should be zero at the maximum driving force, since the reactive driving force diagrams are concave with a unique maximum. However, since our discretization interval was 0.05, we have obtained a nonzero minimum value which can be improved by further decreasing the discretization interval. Nonetheless, a unique minimum is justified which, along with Algorithm 4s (section 2 in the Supplementary material), verifies that the design at the maximum driving force has the least sensitivity of the controlled variables to the disturbances; and the highest sensitivity of the controlled variables to the manipulated variables.

4.2 Step 3: Verification through simulation and analysis

Step 3a - Control structure verification: In order to verify the control structure, the linearized model of the process, i.e., the state-space model is obtained and the steady-state gain matrix is constructed for the 2×2 system. To verify the extent of loop interactions, Algorithm 5s (section 2 in the Supplementary Material) is applied to generate the relative gain array (RGA), to calculate the Niederlinski index (N_I), and to check the system stability, eigenvalue analysis of the RD system models both for open loop and closed loop system is carried out. The results are given in Table 5. It is found that the loop interactions are minimal since diagonal values are close to unity ($N_I \approx 1$) and thereby the control schemes determined for the six case studies are checked.

Step 3b - Dynamic open-loop and closed-loop analysis: In order to evaluate dynamic open-loop and closed-loop performances, Algorithm 6s (section 2 in Supplementary Material) is applied. The disturbance scenarios are presented in Table 6 which are introduced through the feed and expressed in terms of the percentage of feed flow rate. For all six case studies, at first, the open-loop responses of the systems to the disturbances listed in Table 6 have been determined.

[Table 5]

[Table 6]

PI controller: The PI controllers are optimally tuned in ASPEN PLUS dynamics using IMC rules⁷¹. In addition to the controller pairings (see Table 10s), the distillate drum level and sump level are controlled by manipulating the distillate and bottom flow rate respectively. For the level controllers, proportional (P) type is employed, and they are included in the model equations to maintain the dynamic model's consistency and stability. Furthermore, a perfect pressure control is assumed, that is, pressure changes in the column are neglected.

MPC: The A , B , and C matrices for a standard continuous-time state-space model are obtained for the integrated design-control solution at the maximum driving force. D matrix is zero. For a linear system with p inputs, q outputs, and n state variables, $A^{n \times n}$ is the system matrix, $B^{n \times p}$ is the input matrix, $C^{q \times n}$ is the output matrix. Using the linear model of the process, an MPC controller is implemented to perform the closed-loop simulation on the designed RD column (plant). The plant inputs are: the condenser duty - Q_c , the reflux mass flow rate - R (kg/hr), the reboiler duty - Q_R , the distillate mass flow rate - D (kg/hr), the bottoms mass flow rate - B (kg/hr), and the feed molar flow rate (kmol/hr). The plant outputs are: the column pressure (stage 1) - P (atm), the mole fraction of the distillate of interest - x^D , the mole fraction of the bottoms of interest - x^B , the reflux drum liquid level - R_{lev} (m), and the sump liquid level - S_{lev} (m). A linear MPC with MIMO control strategy is obtained. The weights on the controlled outputs are associated in such a way that the top and bottom compositions of interest received more relative importance to stay at their nominal set-points. The prediction horizon is set to 30 minutes (large enough to be insensitive to further increases in the prediction horizon), and the control horizon is set to 4 minutes (small enough to not be significantly computationally expensive).

Analysis of results: The open-loop and closed-loop performance for the integrated design-control solution are highlighted in Figures 4-6 for case study 3. For all other case studies, the open-loop and closed-loop performance are shown in Figure 9s-23s (section 8 in the Supplementary Material). The control structure performances for the PI controller and the MPC are highlighted for both disturbance 1 (+ 10% step change in methanol feed flow rate) and 2 (+ 10% step change in acetic acid feed flow rate). Figure 4 shows the open-loop responses of the controlled variables. Figure 5 shows the closed-loop responses of the controlled variables (product compositions at

the top and bottom) as well as the manipulated variables (reflux and reboiler rates). Figure 6 compares the performances of the PI controller with MPC.

It can be noted from Figures 5-6 that the closed-loop responses with both PI controller and MPC corresponding to the IDC-MDF, are able to reject the disturbances and restore the controlled variables to their original set-points with relatively small efforts in the manipulated variables in both the top and bottom control loops. This is also expected since the RGA matrix has values close to unity in the diagonal and close to zero in the off-diagonal, implying minimal interactions between the control loops and facilitating efficient disturbance rejection.

[Figure 4]

[Figure 5]

[Figure 6]

Step 3c - Final selection: The values of the six terms of the multi-objective performance function (see Eq. 2) are calculated for all the case studies. The results for case study 3 are given in Table 7, while Table 11s (section 8 in Supplementary Material) lists the results for all the other case studies. Note that in Table 7, the performance function terms for case study 3 are calculated for disturbances 1 and 2, which are +10% step in the methanol feed flow rate and +10% step in the acetic acid feed flow rate, respectively (see Table 6).

4.3 Comparison with RD design not at the maximum driving force

Further analysis of the RD column with IDC-MDF is made by comparing its performance with an alternative RD column design not corresponding to the maximum driving force. For the purpose of comparison, the number of stages is kept the same for all cases. Feed locations,

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reflux, and boil-up ratios are varied and the same control structure and controlled variables as the ones in the maximum driving force design are used for the consistency of comparison. Both PI and MPC controllers are used for closed-loop simulation for all the alternatives. Details of the alternative designs for all the cases are given in Table 8. The design targets for all case studies are satisfied. The RGA and N_I values for the alternative designs are given in Table 12s (section 8.2 in the Supplementary Material). For all the case studies, the closed-loop performance under PI and MPC controllers for all the alternatives are evaluated for the disturbance scenario given in Table 6. The performance of the controllers is compared in terms of dynamic responses as well as the control actions for case study 3 at the alternative design. For other case studies, the results are shown in Figure 24s-33s (section 8.2 in the Supplementary Material). For case study 3, Figure 7 shows the transient responses of the controlled variables (product compositions at the top and bottom) as well as the manipulated variables (reflux and reboiler rates), while, Figure 8 compares the performances of the PI controller with MPC for the alternative RD design. It can be noted from Figures 7-8 that compared to Figures 5-6, the controllers are not able to efficiently restore all the controlled variables to their set-points. Furthermore, a relatively longer settling time is observed in the controlled variables. It was noted that for the alternative designs, the diagonal values of the RGA deviate further from unity, compared to IDC-MDF, requiring the controllers for the alternative designs to cope with a larger degree of loop interactions. Thus, it further verifies that moving away from the maximum driving force results in a more difficult control and/or operation of the RD column.

Finally, the multi-objective performance function values for both the IDC-MDF and the alternative design under both the PI and MPC controllers for case study 3 under both disturbance

scenarios are given in Table 7. Table 13s and 14s (section 8.2 in the Supplementary material) list the performance function values for the other case studies.

[Table 7]

[Table 8]

[Figure 7]

[Figure 8]

5. Conclusion

The enhanced controllability of a design obtained through a decomposition based integrated design-control framework using the driving force concept has been highlighted. The framework has been applied to study six RD systems with different configurations. A multi-objective performance function that takes into account the steady-state sustainability metrics (total energy consumption, total carbon footprint) and dynamic control performance (total variations in manipulated variables, the integral absolute error in controlled variables) has been employed. The results show that the RD systems designed at the maximum driving force can be more easily controlled with less error through the PI controllers as well as the MPC controller. For each RD system, the control performance at this operating point is found to be better than other designs, which are not based on the maximum driving force. For a double feed, multi-element system (methyl-acetate production), it has also been demonstrated that the PI and MPC control structures can successfully reject the disturbances introduced through either of the two feeds for the integrated design-control solution. Therefore, it can be concluded that the design at the maximum driving force facilitates the control structure design and results in better controller performance, regardless of the choice of the controller (MPC or PI), compared to designs not

corresponding to the maximum driving force. Current and future work will further extend the framework together with the toolbox with additional integration features and computer-aided tools to enable efficient, reliable, and systematic studies of a wider range of RD systems, including integrated design-control of dividing wall column systems with or without reactions. This way, the overall goal of establishing the RD systems as intensified process alternatives that can be designed to be both sustainable and controllable, would be achieved. Also, the goal is to extend this work to the optimization of design-control under uncertainty, considering also the weights for the different terms of the multi-objective performance function.

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Table 1: Common variables for design and control.

Variables	Role in design	Role in control
Product composition (x^D, x^B)	Design specification (constraint) - provides the setpoint for the controlled variable	Controlled variable- the controller maintains the RD column at the set point
Reflux ratio (RR)	Design variable (calculated from the available driving force diagram) – provides the reference value for the actuator	Manipulated variable- adjusted to control top product composition
Reboiler duty (Q_R)	Design variable (calculated from the available driving force; determines the carbon footprint of the process) – provides reference for actuator	Manipulated variable- adjusted to control bottom product purity
Light key / equivalent light key element composition (D_x) at the maximum driving force	Values for design variables (reflux ratio/rate and/or reboil ratio) are calculated at the located maximum driving force together with the feed location ¹² as, $N_F = N(1 - D_x)$	Sensitivities of controlled variables with respect to disturbance, and the manipulated variables with respect to controlled variables are calculated analytically to check controllability at the maximum driving force ⁵³

Table 2: Key information for problem formulation (step 1).

Case study	Reactions	Catalyst	Number of elements	Main design target	Reference
1	isobutene + methanol $\rightleftharpoons$ MTBE	Sulfuric acid	2	MTBE > 80% at bottom	12, 63
2	isobutene + methanol + 1-butene $\rightleftharpoons$ MTBE + 1-butene	Acidic catalyst	3	MTBE > 90% at bottom	12, 63
3	MeOH + HOAc $\rightleftharpoons$ MeOAc + Water	Sulfuric acid	3	MeOAc > 99% at distillate	61
4	2 Toluene $\rightleftharpoons$ Benzene + <i>o</i> -xylene	zeolite	2	Benzene > 97% at distillate	12, 64
5	isobutene + ethanol $\rightleftharpoons$ ETBE	Ion exchange resin	2	ETBE > 80% at bottom	65, 66, 67
6	2M1B + methanol $\rightleftharpoons$ TAME	Acid ion exchange resin	2	TAME > 97% at bottom	68, 69, 70

Table 3: The element matrix and element reaction for the multi-element reactive systems.

Case study	Element definition and reaction	Components	Formula Matrix		
			A	B	C
3	$A = \text{CH}_4\text{O} \quad B = \text{C}_2\text{H}_2\text{O} \quad C = \text{H}_2\text{O}$ $A + BC \rightleftharpoons AB + C$	Methanol (CH_4O)	1	0	0
		Acetic Acid ($\text{C}_2\text{H}_4\text{O}_2$)	0	1	1
		Methyl Acetate ($\text{C}_3\text{H}_6\text{O}_2$)	1	1	0
		Water (H_2O)	0	0	1

Table 4: Reactive distillation design parameters at the maximum driving force.

Case studies	Number of stages	Feed location stage		Reflux ratio	Reboiler duty (kW)	Condenser duty (kW)
1	6	2		3.88	421.22	1296.11
2	7	4		2.81	427.53	530.80
3	45	Feed 1	Feed 2	1.86	3108.33	5244.44
		7	12			
4	13	6		7.51	930.98	2961.04
5	6	3		3	818.11	729.65
6	9	4		2.63	246.13	1248.96

Table 5: RGA, Niederlinski index and system stability for design at the maximum driving force.

Case studies	Relative Gain Array (RGA)	Niederlinski index (N_I)	Real parts of all eigenvalues negative? (both open loop and closed loop)	Stability
1	$\begin{bmatrix} 1.26 & -0.26 \\ -0.26 & 1.26 \end{bmatrix}$	0.79	Yes	Stable
2	$\begin{bmatrix} 1.26 & -0.26 \\ -0.26 & 1.26 \end{bmatrix}$	0.79	Yes	Stable
3	$\begin{bmatrix} 1.09 & -0.09 \\ -0.09 & 1.09 \end{bmatrix}$	0.91	Yes	Stable
4	$\begin{bmatrix} 0.85 & 0.15 \\ 0.15 & 0.85 \end{bmatrix}$	1.21	Yes	Stable
5	$\begin{bmatrix} 0.98 & 0.02 \\ 0.02 & 0.98 \end{bmatrix}$	1.03	Yes	Stable
6	$\begin{bmatrix} 1.10 & -0.10 \\ -0.10 & 1.10 \end{bmatrix}$	0.77	Yes	Stable

Table 6: Disturbance scenarios

Case studies	Disturbance scenarios	
1	+10% step change in isobutene flow rate.	
2	+16.5% step change in methanol flow rate.	
3	Disturbance 1	Disturbance 2
	+10% step change in methanol	+10% step change in acetic acid
4	+5% step change in Toluene flow rate.	
5	+10% step change in isobutene flow rate.	
6	+10% step change in 2M1B flow rate.	

Table 7: Summary of the performance function terms for the IDC-MDF and the alternative designs for PI and MPC control structure for case study 3.

Objective Function Parameter	Disturbance Scenario 1		Disturbance Scenario 2	
	Design-control solution	Alternative 1	Design-control solution	Alternative 1
J_1 [kW]	8355.26	14135.13	8355.26	14135.13
$J_{2,D}$ [hr] (x_{MeAc}^D by R) (PI)	7.8E-05	10.1979	1.67E-04	0.0116
$J_{2,D}$ [hr] (x_{MeAc}^D by R) (MPC)	0.0062	0.0454	0.0126	0.0055
$J_{2,B}$ [hr] (x_{water}^B by Q_R) (PI)	0.004	0.1145	0.025	0.0123
$J_{2,B}$ [hr] (x_{water}^B by Q_R) (MPC)	6.09E-04	0.1284	0.0042	0.0105
$J_{3,D}$ [-] (PI)	1.57E+03	7.86E+04	3.2E+03	5.04E+03
$J_{3,D}$ [-] (MPC)	2.1E+04	9.43E+04	2.26E+04	4.28E+04
$J_{3,B}$ [-] (PI)	0.718	33.2587	2.007	2.4028
$J_{3,B}$ [-] (MPC)	11.4826	36.92	15.64	25.4562
J_4 [-]	$\begin{bmatrix} 1.09 & -0.09 \\ -0.09 & 1.09 \end{bmatrix}$	$\begin{bmatrix} 0.14 & -0.86 \\ -0.86 & 0.14 \end{bmatrix}$	$\begin{bmatrix} 1.09 & -0.09 \\ -0.09 & 1.09 \end{bmatrix}$	$\begin{bmatrix} 0.14 & -0.86 \\ -0.86 & 0.14 \end{bmatrix}$
J_5 [-]	0.911	7.288	0.911	7.288
J_6 [CO ₂ eq per kg feed]	0.197642	0.317962	0.197642	0.317962

Table 8: Summary of alternative designs for the six case studies

Case studies	Number of stages	Feed stage		Reflux ratio	Reboiler duty (kW)	Condenser duty (kW)
1	6	3		5	616.69	1490.57
2	7	2		7	1055.74	1162.44
3	45	9	12	6.28	6075.34	8059.79
4	13	8		9	1448.46	3478.34
5	6	4		3	823.797	740.773
6	9	3		3	369.664	1370.41

Figure 1: Integrated RD process design-control framework (Algorithm numbers marked with ‘s’ indicates they are included as Supplementary Material). Each box of data-flow indicates data that goes to a step and the generated data that comes from a step; the data stored is cumulative.

Figure 2: Process-controller block diagram highlighting the PI and MPC controller.

Figure 3: Calculated reactive driving force diagram for the 6 reactive systems; A) MTBE without inert (1 atm); B) MTBE with inert (11 atm); C) Methylacetate (1 atm); D) Toluene disproportionation (1 atm); E) ETBE (15 atm); F) TAME (4 bar).

A

B

Figure 4: Open-loop response of the IDC-MDF to step change in feed flow rate; A) +10% methanol; B) +10% acetic acid.

A

B

Figure 5: Closed-loop response of the IDC-MDF for a step change in the feed flow rate; A) PI controller, +10% methanol; B) PI controller, +10% acetic acid; C) MPC controller, +10% methanol; D) MPC controller, +10% acetic acid.

Figure 6: Comparison between the controlled outputs under the MPC and PI controllers for IDC-MDF; A) +10% methanol; composition B) +10% methanol; percent deviation; C) +10% acetic acid; composition D) +10% acetic acid; percent deviation.

Figure 7: Closed-loop response of the alternative design for a step change in the feed flow rate; A) PI, +10% methanol; B) PI, +10% acetic acid; C) MPC, +10% methanol; D) MPC, +10% acetic acid.

Figure 8: Comparison between the controlled outputs under the MPC and PI controllers for the alternative design; A) +10% methanol; composition B) +10% methanol; percent deviation; C) +10% acetic acid; composition D) +10% acetic acid; percent deviation.