

# Active constraint regions for optimal operation of chemical processes - Part I: Application to a reactor-separator-recycle system

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## Abstract

When selecting how to control a chemical plant, with optimal operation in mind, it is important to know the active constraints. It is particularly important to know how the active set of constraints changes with disturbances. In this paper, we study optimal operation of a reactor-separator-recycle process. We seek to find how the set of active constraints changes with varying feed flow rate and energy cost. We explain how to draw a map of active constraints over a two-dimensional disturbance space by a combination of process insight and solving of nonlinear optimization problems. The goal is to sketch the active constraint regions sufficiently accurately, while solving as few optimization problems as possible. For the case study process, we find five distinct regions with different active sets, as well as the highest feed rate the system can process.

**Keywords:** Optimal operation, active constraint regions, disturbances, reactor-separator-recycle

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# Introduction

## Motivation

In optimization and control of chemical processes, active constraints play an important role. The set of active constraints influence both on plant economy and plant control. If one does not control the active constraints, one will have to accept economical loss.<sup>1</sup> Also, knowing which variables are optimally at their constraint value is crucial for control structure design. If a manipulated variable which is optimally at a constraint, is used to control another variable, we can not keep it at its optimal value at all times. Thus it is necessary to know how the active constraints vary with disturbances. It is also useful to know which active constraints are important for plant economy and which are not. Say, for example, that a variable is optimally unconstrained in a particular region, and we decide to use this variable for control. Now if this variable is constrained in a neighboring region, we may have to replace it in our control scheme. However, if backing off from the constraint gives a *small* loss, it might be acceptable to keep the same control structure.

Reactor-separator-recycle systems have been researched actively over the years and is a widely used example in control literature.<sup>2,3,4,5,6</sup> These articles mainly focus on basic regulatory control. Little work has gone into identifying economically optimal operating conditions for such systems, though. Larsson<sup>7</sup> focuses on choosing *self-optimizing variables*<sup>8</sup> among the unconstrained variables. Maarleveld and Rijnsdorp<sup>9</sup> offer a study of constraint control on distillation columns, and discusses how constraints change with different process variables. Except for<sup>9</sup> and the recent work by Jagtap et.al.,<sup>10</sup> there is little emphasis on active constraints in the literature.

## Contribution and organization of the paper

When one seeks to find the (approximate) regions in which each constraint is active, one will usually have to carry out at least a few optimizations. In multi-parametric programming one will typically carry out *many* optimizations, and if the optimization problem itself is difficult, this may be a time-consuming process. In this paper, we address how one can use knowledge about the

optimization problem and process model to simplify this work, and obtain approximate active constraint regions using few optimizations. Part of the purpose is to explain what we can find out just using process knowledge, and what we must solve for numerically. We suggest a step-by-step method for sketching the active constraint regions for a two-dimensional disturbance space, and use this method on a reactor-separator-recycle process. The paper is structured as follows:

- First, we briefly discuss optimal operation in general and discuss the link between the input space and the disturbance space.
- Then we include some general points about optimization to provide background for the rest of the paper.
- We outline a method for finding approximate active constraint regions.
- We describe the process we have used for the case study
- We define optimal operation of the process and provide a degree-of-freedom analysis
- We give the results of the case study
- Then we discuss the results and the efficiency of the method, and we compare our results to those from.<sup>10</sup>
- Finally we give conclusions and suggestions for future work.

## **Optimal operation and constraint regions**

When optimizing operation of chemical processes, we start with formulating a cost function  $J$  which is to be minimized, and identifying the degrees of freedom  $u$  that can be adjusted to minimize  $J$ . Since we consider steady-state optimization, causality is not an issue - thus it does not matter which variables we select as our degrees of freedom, as long as the remaining variables of the model form an independent set. For example, when optimizing a distillation column, the degrees

of freedom could be flows (e.g. reflux  $L$  and vapor boilup  $V$ , so  $u = [LV]$ ) or compositions ( $u = [x_B x_D]$ ). Finally, we need to identify the constraints  $c$  on operation (for example  $L, V > 0, V \leq V_{max}$ ), and the most important disturbances  $d$ .

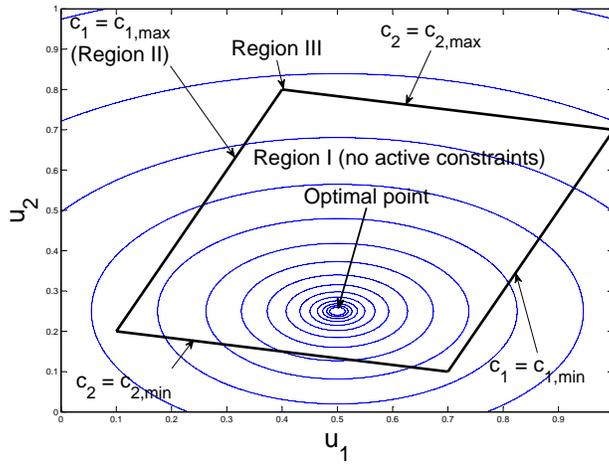
The optimal solution will often be at the intersection of constraints. In Figure 1, we illustrate how constraints and objectives may change when disturbances change, for a simple example with the constraints

$$\begin{aligned}
 c_1 &\geq c_{1,min} \\
 c_1 &\leq c_{1,max} \\
 c_2 &\geq c_{2,min} \\
 c_2 &\leq c_{1,max}
 \end{aligned} \tag{1}$$

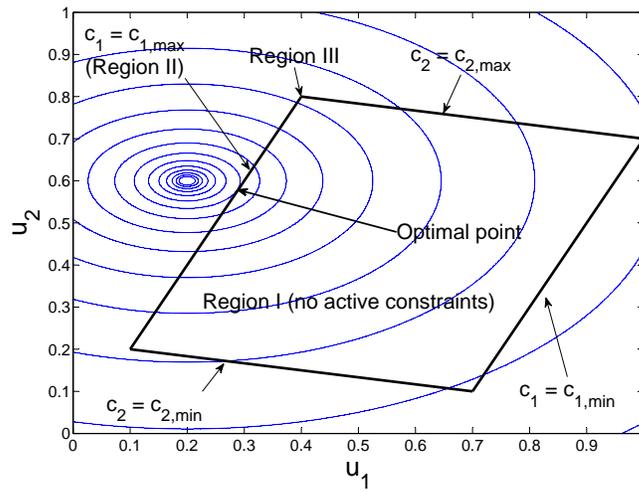
and two degrees of freedom  $u_1$  and  $u_2$ . The constraints define a two-dimensional region in the input space, and the optimal solution may lay in the interior of this region (Figure 1(a)), on one of its edges (Figure 1(b)) or at one of its corners (Figure 1(c)). Operation outside this region is infeasible. In this example the feasible set of  $u$  is shown as being unchanged as  $d$  changes. This will typically be the case if the disturbance which changes is a price. If it is a process disturbance, the constraint lines are likely to change as well.

In this paper, we focus on how the set of active constraints depends on the *disturbances*. The different points indicated in Figures 1(a) - 1(c) would belong in different *constraint regions* in the disturbance space. In the case of one disturbance, each region corresponds to a line segment, as shown in Figure 2. It is worth noticing that "region III" in Figures 1(a) - 1(c) refers to just one point, whereas in the disturbance space, "Region III" includes all  $d$  which makes that particular point optimal.

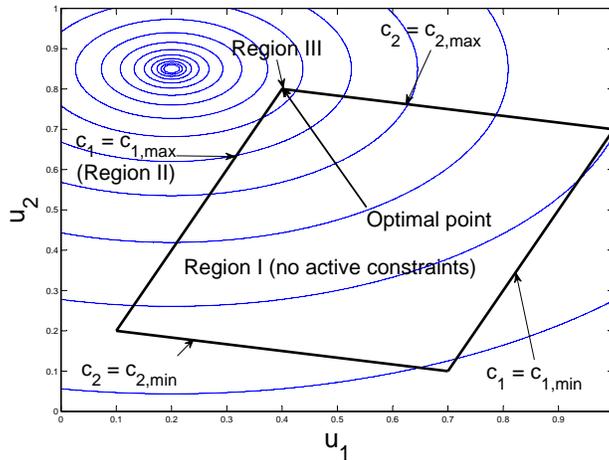
In the case of *two* disturbances, each region is a subset of the now two-dimensional disturbance space. Later in this paper, we use a two-dimensional example to illustrate the suggested method for identifying the active constraint regions. Figure 4(a) is thus a two-dimensional equivalent to Figure 2.



(a) Optimum lies in unconstrained region (I)



(b) Optimum at  $c_1 = c_{1,max}$  (region II)



(c) Optimum at  $c_1 = c_{1,max}$ ,  $c_2 = c_{2,max}$  (region III)

Figure 1: Constraint lines and objective function as functions of degrees of freedom  $u$

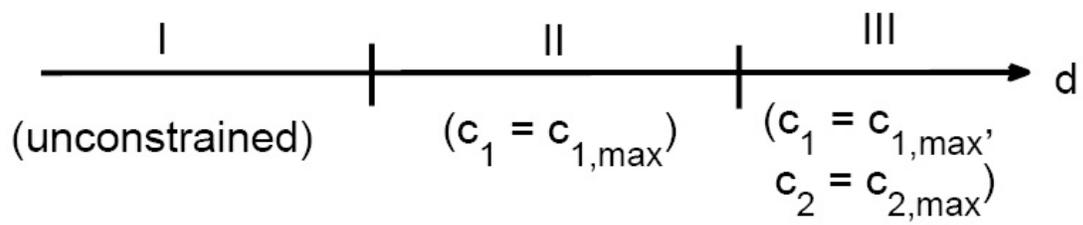


Figure 2: Constraint regions in one-dimensional disturbance space (one disturbance)

# Optimization theory

## General form of the optimization problem

Optimization of chemical processes is typically a nonlinear problem on the form

$$\begin{aligned} & \underset{u}{\text{minimize}} && J(x, u, d) \\ & \text{subject to} && f(x, u, d) = 0 \\ & && c(x, u, d) \leq 0 \end{aligned} \tag{2}$$

where  $J$  is the economical objective,  $f(x, u, d)$  the process model equations and  $c(x, u, d)$  the process constraints. The process model equations may be included in the optimization problem, or solved separately. The latter gives rise to different (and more complex) shapes of  $J$  and  $c$ , but eliminates  $x$  from the equations. Which approach to use depends on which is more robust - i.e. which approach is most certain to return a result. Especially in cases where recycles are involved, it may be beneficial (or even necessary) to let the optimization solver solve the flowsheet model as well (for a discussion, see<sup>11</sup>).

A solution of such a nonlinear optimization problem is characterized by the *Karush-Kuhn-Tucker conditions*<sup>12</sup>). These are as follows (with  $X$  including both  $x$  and  $u$ , but not the disturbance  $d$ ):

$$\begin{aligned} \nabla_X \mathcal{L}(X^*, \lambda^*) &= 0 \\ c_i(X^*) &= 0 && \text{for } i \in \mathcal{E} \\ c_i(X^*) &\leq 0 && \text{for } i \in \mathcal{I} \\ \lambda_i^* &\geq 0 && \text{for } i \in \mathcal{I} \\ \lambda_i^* c_i(X^*) &= 0 && \text{for } i \in \mathcal{E} \cup \mathcal{I} \end{aligned} \tag{3}$$

The optimal solution  $(X^*, \lambda^*)$  is parametrized by the disturbance  $d$ . We seek to find the disturbance value for which a constraint switches from active to inactive - let us call this value  $d_{active}$ .

Here, we take advantage of the fact that for any constraint  $c_i$ , either the constraint value itself or its corresponding Lagrange multiplier  $\lambda_i$  (or both) is zero, and that the sum of the two must be monotonous at least in a small area around  $d_{active}$ . At  $d_{active}$ , we have that  $c_i + \lambda_i = 0$ . Thus, finding  $d_{active}$  is equivalent to solving the equation

$$s_i(d) = c_i(d) + \lambda_i(d) = 0 \quad (4)$$

for  $d$ . Since the optimal solution  $X^*(d)$  is the result of an iterative process, one can not use an analytic method to solve for  $s_i = 0$ , thus an interpolation method is the simplest option. When using MATLAB, this will typically be the **fzero.m** solver. When using this solver, one needs to give two initial points between which the solver should search for the solution. The sign of  $c_i + \lambda_i$  must be different at the two initial points in order for the solver to work.

## Significance of Lagrange multipliers for active constraints

Knowing the active constraint regions allows us to design a control structure based on the expected disturbances. However, even though we may find an optimal control structure for each region, we may be interested in simplifying it, for example by using the same control structure in several regions even if it is not optimal. If using a control structure different from the optimal one, we may end up having to back off from a constraint which is optimally active. We may also have to back off from active constraints due to dynamic reasons (for example, controller overshoot); see Figure 3. This gives rise to a loss, and in a small region the magnitude of the loss ( $|\Delta J|$ ) relates to the Lagrange multiplier as follows:<sup>12</sup>

$$|\Delta J| = \lambda_i |\Delta c_i| \quad (5)$$

where  $|\Delta c_i|$  is the distance from the active constraint  $c_i$  (corresponding to "back-off" in Figure 3) and  $\lambda_i$  is the corresponding Lagrange multiplier. What this means, is that if we back off from the active constraint by a small margin  $\Delta c_i$ , we will have a loss which is large when the corresponding

Lagrange multiplier is large, and small otherwise. Thus, the Lagrange multiplier tells us how hard we get punished by backing off from a constraint. Obviously, we get punished harder if we back off from a constraint when we are far from a region where it is inactive, than if we back off from the same constraint at a point where it becomes active. In economics,  $|\Delta J|$  is called a *shadow price*,<sup>13</sup> in optimal control theory we have the related concept of *costate equations*.<sup>14</sup>

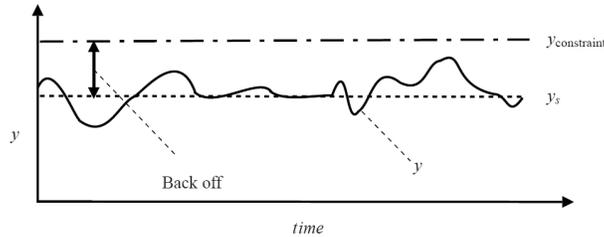


Figure 3: Back off from constraint due to imperfect control (illustration taken from<sup>1</sup>)

## Method for finding active constraint regions as function of disturbances

In this section, we consider the two-dimensional case (with two disturbances) unless otherwise stated. We start this section by defining the terms *constraint line* and *region*:

**Definition 1** *Constraint line: The constraint line corresponding to a constraint  $c$ , is the line separating the regions where  $c$  is optimally active from the regions where  $c$  is optimally inactive.*<sup>1</sup>

**Definition 2** *Region: In this paper, a region refers to a part of the disturbance space, bounded by constraint lines, and described by which constraints are active within that region.*

With the definitions in mind, we can make some general points about how the constraint lines and regions will behave:

<sup>1</sup>In an N-dimensional disturbance space, we will have (N-1)-dimensional constraint surfaces instead.

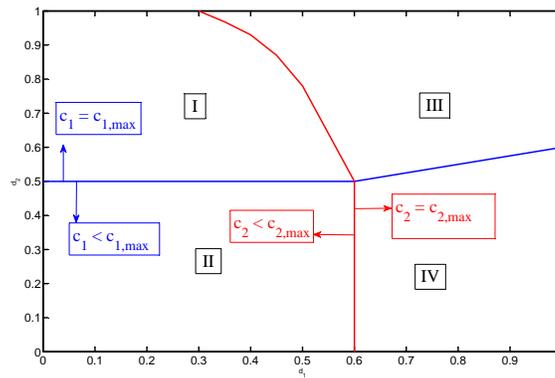
- Constraint lines may cross each other, so one constraint line may span the border between more than two regions. Where two constraint lines cross each other, four regions will meet in a point.
- When one constraint line crosses another, it will generally change slope. This is because the nature of the optimization problem will change when the set of active constraints changes.
- When there are  $N$  constraints, which all may be either active or inactive, there may be as many as  $2^N$  *active constraint sets*. In simple cases, where constraint lines do not cross each other more than once, this means we also have a maximum of  $2^N$  regions (as illustrated by Figure 4(a), where we have two constraints and four regions). In more complex cases, where two constraint lines may cross each other more than once, we may have more than  $2^N$  regions. In that case, some regions will share the same active set. However, the number of regions is usually smaller than  $2^N$ , since some constraint combinations may not occur, for example because they are physically impossible. For example, we often have both maximum and minimum constraints on the same variable - these will obviously never be active at the same time.
- In the one-dimensional case (i.e. one disturbance), each region corresponds to a line segment on the disturbance axis (x-axis). This is illustrated in Figure 4(b). Here, the y-axis is used to plot the optimal value of the constraint functions as functions of a single disturbance.

We now want to outline a method for identifying active constraint regions without having to optimize at a large number of points across the whole disturbance space. As an illustrative example, let us consider a hypothetical problem with two constraints:

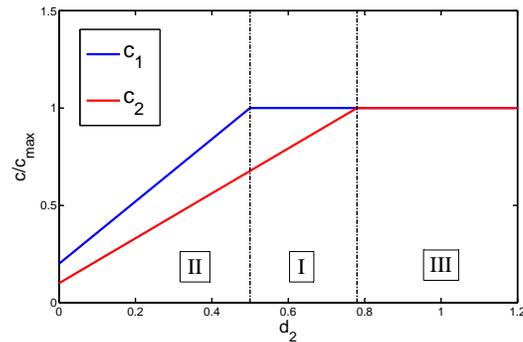
$$[c_1 \ c_2] < [c_{1,max} \ c_{2,max}] \quad (6)$$

We have two disturbances,  $d_1$  and  $d_2$ . Both  $c_1$  and  $c_2$  are continuous functions of the disturbances, and we have four regions:

1. Both constraints are inactive
2. (only  $c_1$  is active)
3. (only  $c_2$  is active)
4. (both constraints are active)



(a) Two-dimensional case (two disturbances)



(b) One-dimensional (one single disturbance)

Figure 4: Example figure illustrating how constraint lines divide the disturbance space into regions, and how the regions translate to line segments in the one-dimensional case

The regions are shown in Figure 4(a): Constraint  $c_1$  is active ( $c_1 = c_{1,max}$ ) below the blue constraint line, and  $c_2$  is active ( $c_2 = c_{2,max}$ ) to the right of the red constraint line. We also show the one-dimensional case where  $d_1 = 0.5$  and  $d_2$  is on the x axis (Figure 4(b)). Using this example as

an illustration, we now outline a method for finding active constraint regions. First it is reasonable to make the following assumptions:

1. In a two-dimensional plot with  $(d_1, d_2)$  along the axes, two neighboring regions will only differ by one active constraint, except when two constraint lines cross each other (in Figure 4(a) this happens in the point  $(0.6, 0.5)$ ). For the opposite to be true, two constraint lines would have to follow each other exactly (at least along a segment).
2. We assume that the same set of active constraints does not appear in two separate regions of the disturbance space (thus, two constraint lines will not cross each other twice).
3. We assume that there will always be a maximum feed rate ("bottleneck") above which we can not satisfy all constraints.

Based on the above definitions, considerations and assumptions, we suggest using the following procedure to find active constraint regions:

1. Using knowledge about the process model and the optimization problem, find out if any constraints will be active (or inactive) for *all* values of the disturbances, thus reducing the number of possible regions by a factor of 2 for each constraint which is always active.
2. Also use the same insight to predict whether some region borders (part of constraint lines) will be independent of one of the disturbances. In a 2D graph, these borders will correspond to vertical or horizontal lines. In the example (Figure 4(a)), the constraint line separating regions I and III from regions II and IV is horizontal (independent of  $d_1$ ). The line segment that separates regions II and IV is vertical (notice that this segment is only a part of a constraint line!)
3. Locate the region borders that are found to be vertical or horizontal, by solving for the disturbance value at which the corresponding constraint changes between inactive and active. In Figure 4(a), this corresponds to finding the value of  $d_2$  for which  $c_1$  becomes active.

4. Likewise, find the value of  $d_1$  for which we go from region II to region IV (that is, for which  $c_2$  becomes active, with  $d_2$  lower than the value we found in the previous step), thus locating vertical part of the constraint line for  $c_2$ .
5. Find at least one more point along the line separating regions I and III. This means we need to do find a value of  $(d_1, d_2)$  which makes constraint  $c_2$  switch from active to inactive. By fixing  $d_2$  at a higher value than the one we found in step 3, and solving for the value of  $d_1$  which makes  $s_1 = 0$ , we find a point on this line.
6. In the same manner, find at least one more point along the line separating regions III and IV (with  $d_1$  fixed at a value *higher* than found in step 4).
7. If we are confident the two last region borders are straight lines, or satisfied with it as an approximation, we do not need more points. However, the true borders will often be curved (as the border between regions I and III in Figure 4(a)), and if we want to describe them more accurately, we need to find more pairs  $(d_1, d_2)$ .

We have used this approach successfully in a case study which we will go through in the following sections.

## Description of the example process

In this paper, we consider the simple reactor-separator-recycle process shown in Figure 5:

Fresh feed of A ( $F$ ) is mixed with recycled distillate from the column. The mixture ( $F_r$ ) goes to the reactor, which is a continuously stirred tank reactor (CSTR) with two reactions (Equation 7. The first yields B, which is the desired product, and the second yields C which is an undesired by-product. A real-life example of such a process could for example be a hydrocarbon isomerization process, where cracking occurs as a side reaction (this is quite common in hydrocarbon refining processes).

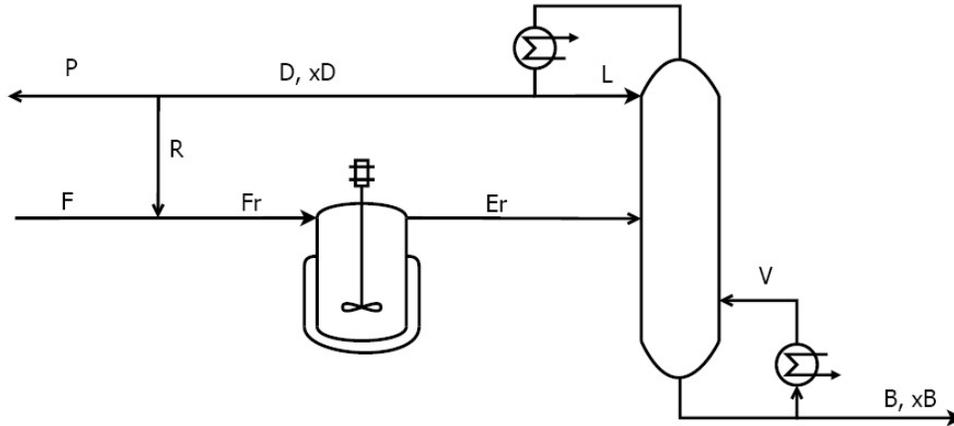


Figure 5: Flow sheet of the reactor-separator-recycle system



In general, the steady state mass balance of a CSTR with one feed stream and one product stream can be written as follows:

$$0 = \frac{dn_r}{dt} = F_r \cdot x_{F,r} + M_r \cdot v \cdot r - E_r \cdot x_r \text{ [mol/s]}
 \tag{8}$$

where  $n_r$  [mol] is the reactor holdup vector,  $F_r$  [mol/s] is the flow rate entering the reactor,  $x_{FR}$  the reactor feed composition,  $E_r$  [mol/s] is the reactor exit flow rate,  $v$  is a matrix of stoichiometric coefficients,  $r = [r_1 \ r_2]^T$  [s<sup>-1</sup>] is a vector of reaction rates and  $x_r$  is the composition inside the reactor, expressed in mole fractions (in a CSTR, the exit stream is assumed to have the same composition as the reactor contents).  $M_r$  [mol] is the total reactor holdup.

We will, for later use, define the reactant conversion  $X$  for the reactant and the product yield  $Y_i$  for each product  $i$ .

$$X = \frac{\dot{n}_{reactant,in} - \dot{n}_{reactant,out}}{\dot{n}_{reactant,in}} \cdot 100\% \quad (9)$$

$$Y_i = \frac{\dot{n}_{i,out} - \dot{n}_{i,in}}{\dot{n}_{reactant,in}} \cdot 100\% \quad (10)$$

where  $\dot{n}$  indicates molar flowrate of a given component.

The reaction model uses first-order kinetics; the reaction rate of reaction  $i$  is given by

$$r_i = k_i x_A \quad (11)$$

where  $x_a$  is the mole fraction of A in the reactor and

$$k_i = A_i e^{-\frac{E_{a,i}}{RT}} \quad (12)$$

where  $E_{a,i}$  is the activation energy of reaction  $i$  (in  $J/mol$ ),  $T$  is the temperature in  $K$  and  $R$  is the gas constant ( $8.3145 \frac{J}{mol \cdot K}$ ). Kinetic data are shown in Table 1. In this example, reaction 1 has a lower rate constant ( $A_1 < A_2$ ), but also a lower activation energy, thus it is the favored reaction at lower temperatures.

Table 1: Reaction kinetics parameters

Reaction	$A$ ( $[s^{-1}]$ )	$E_a$ ( $J/mol$ )
1	$1 \cdot 10^5$	$6 \cdot 10^4$
2	$5 \cdot 10^6$	$8 \cdot 10^4$

The reactor product  $E_r$  is separated in a column. The distillate  $D$  is rich in byproduct (C) and unreacted A, whereas the bottom product  $B$  is rich in the desired product B. For the distillation column, we have used a simple column model using the following assumptions: Constant relative volatilities, constant molar overflow, constant pressure over the entire column, equilibrium at every stage and negligible vapor holdups. Francis' weir equation is used to calculate liquid flow rates. The column data are shown in Table 2.

Table 2: Distillation column parameters

$\alpha_{AC}$	0.70
$\alpha_{BC}$	0.60
Number of stages	30
Feed stage location	15
Feed liquid fraction	1
$V_{max}$	30 mol/s

A fraction of the distillate is recycled ( $R$ ), the remaining distillate leaves the system as a purge stream ( $P$ ).

## Defining optimal operation of the reactor-separator-recycle process

### Steady-state operational economy objective

The objective  $J$  should cover all economical aspects that are influenced by the steady state operation. In general, these include cost of raw materials, energy and utilities (like cooling water), and the value of products. The cost of a product stream may be positive (if the product is sold, or processed to valuable products later on) or negative (if it is waste which must be disposed of). We may generalize to write

$$J = \sum p_{F,i} F_{F,i} + \sum p_{U,j} F_{U,j} + \sum p_{P,k} F_{P,k} \quad (13)$$

where  $F_{F,i}$ ,  $F_{U,j}$  and  $F_{P,k}$  are the flow rates of feeds, utility streams and product streams, respectively - all in mol/s, except for energy usage which is in [\$/kJ].  $p_{F,i}$ ,  $p_{U,j}$  and  $p_{P,k}$  are the prices of the respective streams). In this example, we use the following objective function:

$$J = p_F F + p_V V - p_B B - p_P P \quad (14)$$

where  $F$ ,  $V$ ,  $B$  and  $P$  refer to Figure 5, and the prices are given in Table 3. (The value given for

$p_V$  is listed as variable, it is used as a disturbance later on)

Table 3: Prices used in optimization ( $p_V$  is the nominal value)

Feed	$p_F$	1 \$/mol
Product	$p_B$	2 \$/mol
Purge	$p_P$	0.5 \$/mol
Energy	$p_V$	variable

## Degrees of freedom

For example by using the method outlined by,<sup>15</sup> we find that the process shown in Figure 5 has got six steady-state degrees of freedom. We may also find this number by examining the model, finding that it has 104 variables and 98 independent equations. When using the method from,<sup>15</sup> we relate the six degrees of freedom to:

1. Feed flow rate  $F$
2. Recycle/purge split  $P/D$
3. Reactor holdup  $M_r$
4. Reactor temperature  $T_r$  (since the CSTR has a cooling jacket as shown in Figure 5, we may adjust this)
5. Column reboiler duty  $Q_R$
6. Column condenser duty  $Q_C$

However, in simulation, we may specify *any* six variables as long as the resulting 98-by-98 system is not structurally or numerically singular. When initializing the process model, the following set of specifications was used:

- Reactor temperature  $T_r$
- Feed flow rate  $F$

- Mole fraction of component B in product stream  $B$ ,  $x_{B,B}$
- Reactor holdup  $M_r$
- Column reflux  $L$
- Purge/distillate ratio  $P/D$

## Constraints

When optimizing a chemical process plant, we will encounter the following types of constraints:

- There are always capacity constraints; maximum levels in tanks and liquid-phase reactors, maximum available amount of utilities like steam and cooling water, and maximum feed and product rates.
- There will usually be requirements on product quality, often in terms of maximum content of impurities.
- In addition, there are typically constraints on pressures (due to limits in piping strength) and temperatures (for example to limit catalyst degradation).

In this work, we have included maximum limits on reactor temperature  $T_r$ , reactor holdup  $M_r$  and column boilup  $V$  as well as a minimum limit on the fraction of component B in stream  $B$ . In addition we require that all flow rates are  $\geq 0$ . See Table 4.

Table 4: Constraint values used in optimization

$x_{B,B,min}$	0.90
$T_{r,max}$	390 K
$M_{r,max}$	11000 mol
$V_{max}$	30 mol/s
$R_{min}$	0 mol/s
$P_{min}$	0 mol/s
$B_{min}$	0 mol/s

## Disturbances

In a chemical process, there are many possible disturbances. The most important ones are usually related to feed conditions (flow rate, composition and pressure), as these often depend on the operation of an upstream process. In addition, the prices of feeds, products and utilities are often changing on a daily basis.

We may also have changes in process parameters with time. For example, in a catalytic process the catalyst activity may degrade with time, so that the value of  $A$  in the expression for the reaction rate constant  $k$  decreases. Deposits inside piping may increase pressure drop, and decrease heat transfer. Finally, downstream processes may demand changes in product flow rates and compositions.

In this work, we consider two disturbances:

- Feed flow rate  $F$  (nominal value:  $1.1\text{ mol/s}$ ). This is the flow rate which is most likely to be given by another process unit.
- Energy cost  $p_V$  in the column (nominal value:  $0.01\$/\text{mol}$ ) (either of the four price parameters could be used - what is really important is the energy price in relation to the difference in value between feed and products)

Since the feed flow rate is used as a disturbance, we have only got *five* degrees of freedom in the optimization problem, compared to six degrees of freedom for initialization.

## Results

### Initialization

In order to have a feasible starting point for subsequent optimization, we specified six variables as described above, and used **fmincon** to solve the model equations. To do this, the model equations and specifications were included as equality constraints in the optimization problem (corresponding to  $f(x, u, d)$  in equation 2), and a dummy objective function with a constant value was used.

This approach is used with success in.<sup>16</sup> Table 5 shows the values used for initialization, plus the resulting values of other chosen variables (reactor holdups  $n_{r,i}$ , conversion  $X_A$  and yield  $Y_i$  as well as product and distillate flow rates and compositions). Notice that the specification on  $x_{B,B}$  was set slightly above the minimum value, this was to give an initial solution with a little margin to the most important constraint - on the other hand, the reactor holdup was initialized at its maximum value.<sup>2</sup>

Table 5: Initial data for the reactor-separator-recycle system, used as starting point in optimization.

Variable	Value
$F$	$1.1 \text{ mol/s}$
$x_{B,B}$	0.901
$T_r$	355 K
$M_r$	11000 mol
$L$	$25 \text{ mol/s}$
$P/D$	0.2
$R$	$1.512 \text{ mol/s}$
$n_{r,A}$	5064 mol
$n_{r,B}$	4159 mol
$n_{r,C}$	1777 mol
$X_A$	67.33 %
$Y_1$	61.82 %
$Y_2$	5.51 %
$B$	$0.7649 \text{ mol/s}$
$x_{A,B}$	0.099
$x_{B,B}$	0.901
$x_{C,B}$	0.000
$D$	$1.8901 \text{ mol/s}$
$x_{A,D}$	0.607
$x_{B,D}$	0.167
$x_{C,D}$	0.227

## Active constraint regions

According to the method outlined earlier in the paper, we start by checking if any constraints will be active for all  $(F, p_V)$ . Indeed there are two, namely the constraints on product purity  $x_{B,B}$  and

<sup>2</sup>The reason for having  $T_r = 355 \text{ K}$  despite the maximum is shown as  $T_{r,max} = 390 \text{ K}$  in Table 4 is that we increased the maximum value in order to make the region where the constraint on  $T_r$  was inactive, more visible. Originally  $T_{r,max}$  was 355 K, but this made the region very small.

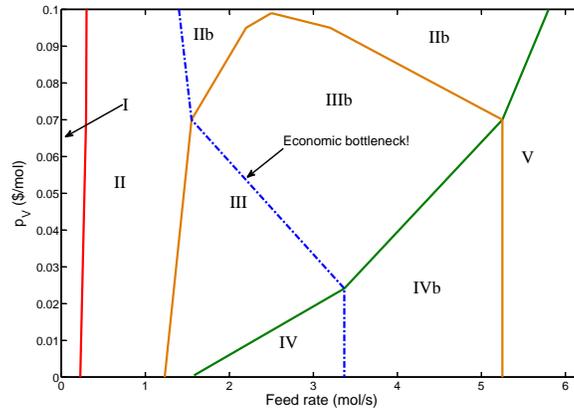
reactor holdup  $M_r$ . The first one follows from the "Avoid Product Giveaway rule", which states that *when the product prices are constant, the minimum purity constraint in the valuable product stream is always active*. The latter is also easy to explain: Reducing the holdup leads to a lower single-pass conversion, yielding more A in the column feed, without improving selectivity in favour of the desired product. Thus the column feed will contain more A and less B, meaning we get less product.

We can also assume that one constraint will never be active - namely,  $P \geq 0$ . This is because as long as C is produced, we need to provide a way out of the system for it. Since C is the most volatile component, we will always have some of it in the distillate, thus we must purge some of the distillate to avoid accumulation of C within the system.

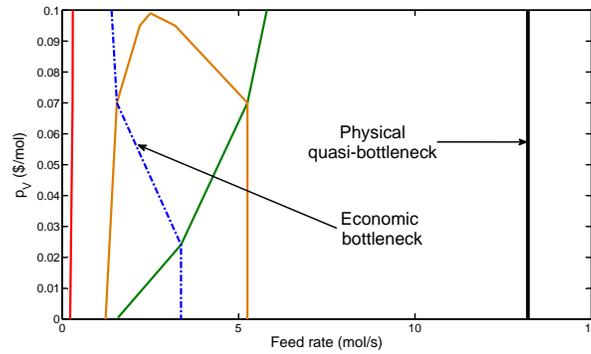
This means we have to find when the following variables are at their constraint values:  $T_r$ ,  $R$ ,  $V$ ,  $D$  and  $B$ . Using **fmincon.m** for optimization and **fzero.m** for interpolation, we come up with the regions shown in Figure 6. Each region is referred to by a number, Table 6 lists which variables are at their constraint value in each respective region. The constraint lines that define the regions are as follows:

- At the red constraint line,  $T_{r,max}$  becomes active.
- Orange constraint line (left part):  $R_{min}$  becomes inactive.
- Green constraint line:  $V_{max}$  becomes active.
- Orange constraint line (right part):  $R_{min}$  becomes active again.
- The dashed blue line could be seen as a "quasi"-constraint line - it indicates where it is not economically optimal to increase  $F$  any more, so it shows the *economic bottleneck* of the process.

In the Discussion, we explain further the shape of the regions. In Table 7, we show the optimal value of selected model variables at four points in the disturbance space. Variables that are at a constraint are shown in **bold**.



(a) Active constraint regions for  $F$  up to  $6\text{ mol/s}$  and  $p_V$  up to  $0.10\text{ \$/mol}$



(b) Expanded version of Figure 6(a) with  $F_A$  up to  $15\text{ mol/s}$ .

Figure 6: Active constraint regions for the reactor-separator-recycle system

Table 6: Constrained variables in each region in Figure 6(a)

Region number	Constrained variables
I	$x_{B,B}, M_r, R$
II	$x_{B,B}, M_r, R, T_r, (F)$
IIb	$x_{B,B}, M_r, R, T_r$
III	$x_{B,B}, M_r, T_r, (F)$
IIIb	$x_{B,B}, M_r, T_r$
IV	$x_{B,B}, M_r, T_r, V, (F)$
IVb	$x_{B,B}, M_r, T_r, V$
V	$x_{B,B}, M_r, T_r, V, R$

Table 7: Optimal values of selected variables for different values of  $F$  and  $p_V$ . Numbers in **bold** indicate active constraints

Region(s)	I	II	IV	IIIb
$F$ [mol/s]	0.1	1.0	3.0	3.0
$p_V$ [\$/mol]	0.02	0.02	0.01	0.07
$F_r$ [mol/s]	0.1000	1.0006	5.4219	3.3829
$E_r$ [mol/s]	0.1024	1.0871	5.6584	3.6079
$T_r$ [K]	328.8	<b>390.0</b>	<b>390.0</b>	<b>390.0</b>
$x_{A,r}$	0.2356	0.0815	0.2229	0.2121
$x_{B,r}$	0.7168	0.7589	0.4604	0.6225
$x_{C,r}$	0.0476	0.1596	0.3167	0.1654
$Y_1$ [%]	73.43	82.50	61.11	68.44
$Y_2$ [%]	2.44	8.64	6.30	7.17
$B$ [mol/s]	0.0781	0.9108	2.3696	2.0511
$x_{A,B}$	0.1000	0.0900	0.0996	0.0996
$x_{B,B}$	<b>0.9000</b>	<b>0.9000</b>	<b>0.9000</b>	<b>0.9000</b>
$x_{C,B}$	0.0000	0.0100	0.0004	0.0004
$D$ [mol/s]	0.0243	0.1762	3.2888	1.5569
$x_{A,D}$	0.6707	0.0375	0.3117	0.3603
$x_{B,D}$	0.1290	0.0300	0.1436	0.2569
$x_{C,D}$	0.2003	0.9325	0.5447	0.3828
$L$ [mol/s]	1.6231	2.1365	26.7112	14.0009
$V$ [mol/s]	1.6475	2.3128	<b>30.0000</b>	15.5578
$P/D$	1.0000	1.0000	0.2636	0.7541
$R$ [mol/s]	<b>0.0000</b>	<b>0.0000</b>	2.4219	0.3829
$J$ [\$/s]	-0.035	-0.863	-1.873	-1.378

# Discussion

## Shape of active constraint regions

In the following, we will explain the presence of each active constraint region, at least for those who are not obvious.

- The dashed blue line in 6(a) indicates where it is no longer optimal to increase production. That is, if  $F$  was a degree of freedom and  $F_{max}$  was specified instead, the blue line shows where the constraint  $F < F_{max}$  would no longer be active. We illustrate this further with Figure 7(a), where the value of the objective function  $J$  at the optimal solution is given as a function of  $F$  at three different energy prices.
- We notice that the right part of the orange constraint line in Figure 6(a), separating regions IVb and V, is vertical. This is easily explained;  $V_{max}$  is active here, and once the optimal value of  $V$  reaches  $V_{max}$ , the next region boundary must be independent of  $p_V$ , thus it is vertical.
- We also see that for very low  $F$ , where no capacity constraints are active, we have a region (region 1 in Figure 6(a)) where the maximum reactor temperature ( $T_{r,max}$ ) is not an active constraint. This is because when the overall conversion is very high (as it is at low flow rates), we benefit from increasing the reaction *selectivity* in favor of the desired reaction, this compensates for the lower overall conversion. As mentioned in the process description, reaction 1 has a lower activation energy, and thus will be favored by low temperatures.
- $R_{min}$  is active ( $R = 0$ ) at low  $F$ : If  $F$  is sufficiently low, the reactor exit stream contains very little unreacted A. Thus there is no benefit from recycling, as we would only be recycling by-product C. If the value of the purge stream was zero, however, we would recycle as long as there is any A in the distillate at all.
- $R_{min}$  is active at high  $F$ : As  $F$  increases, the reactor conversion goes down (Figure 8(a)) and

the product stream will contain more unreacted A. This must be compensated by increasing  $V$  or decreasing  $L$  in the column. If  $V = V_{max}$ , we cannot increase it further and our only option is to reduce  $L$ , meaning the distillate flow rate increases and the bottoms flow rate decreases (see Figure 8(b)). In this situation, we have nothing to earn from recycling more, so the entire increase in distillate flow rate goes into the purge stream  $P$ , leading to a higher purge ratio  $P/D$ . For higher  $p_V$ , we may find the same even when  $V < V_{max}$ , because an increase in  $V$  costs more than it gives. We discuss this further below.

- To the right of the constraint line for boilup rate  $V$ , *all* other constraint lines (including the "quasi-constraint line" for  $F_{max}$ ) are vertical, because when  $V$  is fixed at  $V_{max}$ , the objective function is independent of  $p_V$ , and thus the optimal solution is also independent of  $p_V$ .

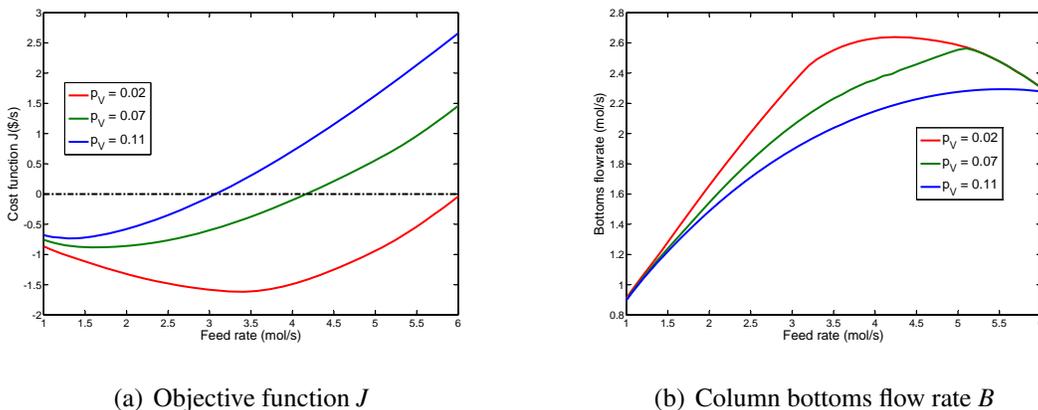


Figure 7: Optimal objective function value  $J$  and column bottom product flowrate  $B$  as function of feed flow rate  $F$  at three values of  $p_V$

That  $M_{r,max}$  is always active, means that  $M_r$  should be controlled at this constant value  $M_{r,max}$ . However,<sup>7</sup> argues that if energy is cheap (i.e.  $p_V$  is small) and  $V_{max}$  is large, it could be beneficial, for control purposes, to let the holdup vary, giving better disturbance rejection.

The "physical quasi-bottleneck" indicated in Figure 6(b) is reached when the optimal value of  $B$  reaches zero: As we increase the feed rate, less of the reactant is converted to products. Thus, in order to satisfy the constraint on  $x_{B,B}$  we need to decrease  $B$  (see also Figure 7(b)) until it can

not be decreased further, by reducing the reflux flow rate  $L$ . The full set of active constraints at the physical quasi-bottleneck is:

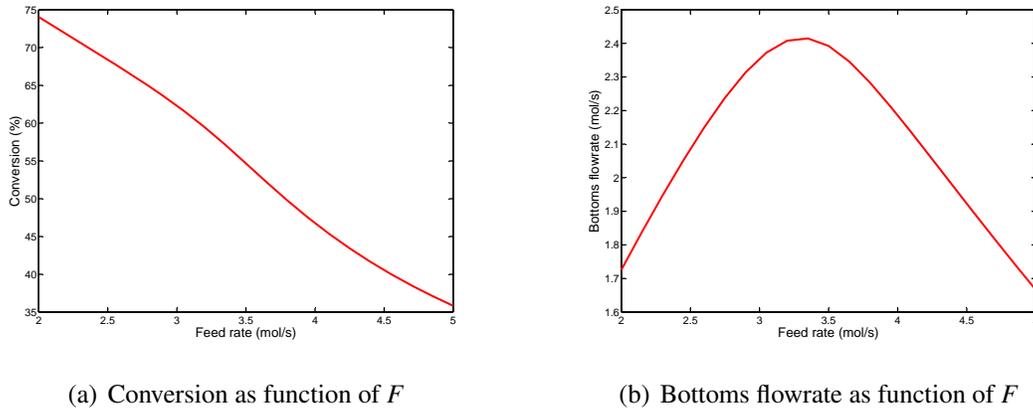


Figure 8: Reactor conversion  $X$  and bottoms flow rate  $B$  as functions of  $F$  when  $M_r$ ,  $x_{B,B}$ ,  $T_r$ ,  $V$  and  $P/D$  are fixed.

$$x_{B,B} = x_{B,B,min}$$

$$M_r = M_{r,max}$$

$$T_r = T_{r,max}$$

$$V = V_{max}$$

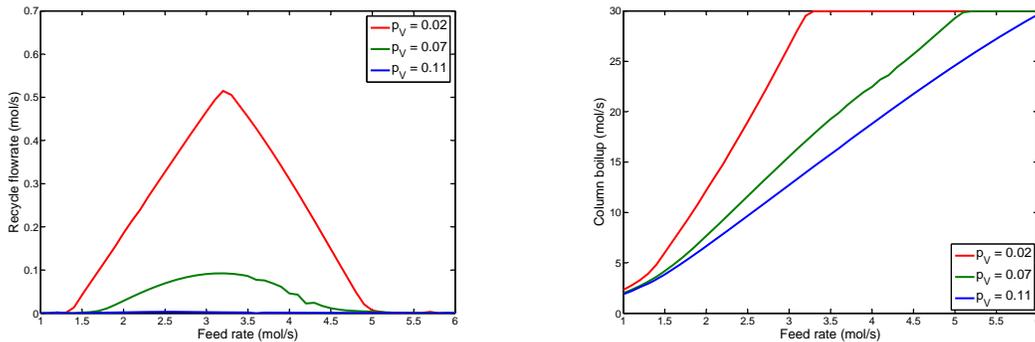
$$B = 0$$

$$R = 0$$

However, it would be possible to take in more feed to the system - we could take in  $V_{max}mol/s$ , and boil up and purge the entire stream. This could for example be necessary if this process was the only way out for the product of an upstream process. This is why we refer to it as a quasi-bottleneck.

## More on the effect of recycling

As mentioned above, we found that for low and high  $F$  it was optimal to purge all the distillate from the column, rather than recycling some of it. In Figure 9(a), we show how the optimal value of  $R$  (the recycle flow rate) varies with  $F$ , and Figure 9(b) shows the same for vapor boilup in the column. As we can see, at the lowest  $p_V$  the maximum recycle flow rate is reached just as  $V$  reaches  $V_{max}$ . However, for the two higher  $p_V$  values, the maximum recycle flow rate is reached *before*  $V$  reaches  $V_{max}$ . The explanation for this is: When we increase the reactor throughput, we get a lower conversion and thus the column feed is leaner in the heavy component  $B$ . Thus, to maintain the desired purity, most of the increased column feed goes out as distillate, *not as valuable bottom product*. Thus, we get less revenue for the increased reactor throughput. At some point, we cannot increase the bottom product flow rate any more, and thus any recycled material would just have to be boiled up again. Then it is more profitable to purge it, and we reach the point where  $R_{optimal} = 0$ . If  $p_V$  is sufficiently high, it will be optimal to have  $R = 0$  for *all*  $F$  - in Figure 6(a) we see that this happens at  $p_V \approx 0.1$  \$/mol.



(a) Optimal value of recycle flow rate  $R$  as function of  $F$  (b) Optimal value of boilup flow rate  $V$  as function of  $F$

Figure 9: Selected variables as function of feed flowrate  $F$  for three different values of  $p_V$

## Efficiency of method

All optimizations were carried out without using analytical Jacobians, so the optimization solver would typically need 50-100 iterations to converge to a solution. However, each iteration was quick since calculation of the objective and constraint function values was easy (as they involve no further equation solving). An interpolation search would typically require about 10 function evaluations (optimizations). If one should find the active constraint regions by just mapping the active constraints at a number of points evenly distributed over the disturbance space, it would be necessary to carry out many more optimizations - even if one kept the assumption that every region boundary was straight.

The fact that the minimum constraint on  $R$  is active for both high and low  $F$ , but not for intermediate, gave rise to an additional challenge. For **fzero.m** to work, it requires two end points where the function for which we seek a zero, has different signs. Since the constraint was active both at high and low  $F$ , choosing values near 0 and  $F_{max}$  would yield two end points where the function value was positive. Thus we had to carry out a few extra optimizations at intermediate  $F$  to find a point where the optimal  $R \neq 0$ .

## Comparison with Jagtap et.al.

<sup>10</sup> study a process where two reactants A and B react according to the following reaction scheme:



with C being the desired product and D being an unwanted byproduct. Instead of a purge stream, they use *two* distillation columns, and the desired product C is taken out in the distillate stream of the second column (however, the bottoms stream of that column may be seen as a replacement for a purge stream as this stream takes care of most of the by-product D). They use

the "avoid product giveaway" rule to fix *three* specifications, all are directly related to exit stream compositions in the distillation columns. However, they use more indirect specifications, namely the ratio of B to C in the bottom stream from the first column, and the *loss* of C and D in the bottom and distillate streams, respectively, in column 2. These seem to be chosen more for the sake of easy steady-state convergence of the process model.

Like in this work, they find that when fresh feed is a degree of freedom, there is a feed rate at which the plant profit reaches a maximum, and any further increase in feed rate leads to a *decrease* in profit. This is because the increased feed rate does not lead to a sufficiently large increase in the amount of valuable product. An increased feed flow rate means a lower conversion percentage, so one gets more of the impurities in the reactor product. Then more needs to be recycled, and the increase in production rate is not high enough to "pay" for the increase in feed consumption rate.

A notable difference is that the region where the reactor temperature constraint is inactive, is much larger, and another constraint becomes active before it ( $V_{max}$  in the first column). This is probably because the reaction parameters are such that the temperature has a much stronger influence on reaction selectivity.

## **Conclusions and future work**

### **Conclusions**

In this paper, we have outlined a method for finding active constraint regions for a chemical process. We have discussed optimal operation of reactor-separator-recycle systems and applied the above mentioned method to describe the active constraints of a reactor-separator-recycle process. We have found that we have 5 distinct active constraint regions, and that there is a flow rate above which we cannot operate without breaking constraints. For the data used in this work, this flow rate is  $13.2\text{ mol/s}$ .

## Future work

After having successfully applied the above mentioned method on distillation columns and a reactor-separator-recycle system, which are both well researched types of processes, we want to apply it to an LNG process.

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