Control of Reactor and Separator, with Recycle

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Abstract

This paper looks at control of a plant that consists of a reactor, separator and recycle of unreacted reactant. This configuration is very common in industry, and includes both liquid phase and gas phase systems. Some examples of gas phase systems are ammonia and methanol plants.

For example, consider a CSTR reactor where component A is converted to a product and the amount converted is given by $k(T)Mz$ $[mole A/s]$. To increase the conversion one then has three options:

1. Increase the temperature which increases the reaction constant k

2. Increase the fraction of A in the reactor, z. This can be done indirectly by increasing the amount of recycle of A.

3. Increase the reactor holdup M.

In a liquid phase system the reactor holdup is determined by the reactor level, and in a gas phase system by the reactor pressure. Here we will assume that the temperature is constant.

Since at steady-state with given product specifications the conversion of A in the reactor is given by the feed rate, it follows that only one of the two remaining variables mentioned above can be controlled independently (or more generally, one variables that influences these variables), and we must let the second variable "float" and adjusts itself.

Two common control strategies are then (A) to keep the reactor holdup constant (and let the recycle flow float) or (B) to keep the recycle flow constant (and let the reactor holdup float). In case (A) one may encounter the so-called "snowball effect" where the recycle goes to infinity. This occurs because at infinite recycle flow we have $z = 1$ which gives the highest possible production. In effect, the snowball effect occurs because the reactor is too small to handle the given feed rate, so it is really a steady-state design problem.

Luyben has studied liquid phase systems in a number of papers, and has concluded that a variant of control strategy (B) (where the reactor level is used as through-put manipulator) should be used to avoid the "snowball effect". However, from an economic point of view one should usually for liquid phase systems keep the reactor level at its maximum value (to maximize the conversion per pass), so Luyben's recommendation has an economic penalty which it seems that most researchers so far have neglected.

However, for the gas phase system the situation is different, there are a cost associated with reactor hold-up (pressure). The optimum are unconstrained in this variable. Fixing recycle rate, purge fraction, or reactor pressure had all good self-optimizing properties. This is linked to the behavior of these variables as conversion increases. As expected purge flow is a bad alternative as a controlled variable. More unexpectedly, inert composition in the recycle turned out to bad self-optimizing properties. This is also explainable by the behavior of this variable when conversion is increased. The results for the gas phase reactor carries well over to the methanol case study.

We support our conclusions by the use of simple models.

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1   Introduction

A common feature of many chemical processes is the presence of recycle. One example of mass recycle is a reactor, separator with recycle of unreacted reactant, see figure 1. This configuration is very common in industry, and includes both liquid phase and gas phase systems. Some examples of gas phase systems are ammonia and methanol plants.

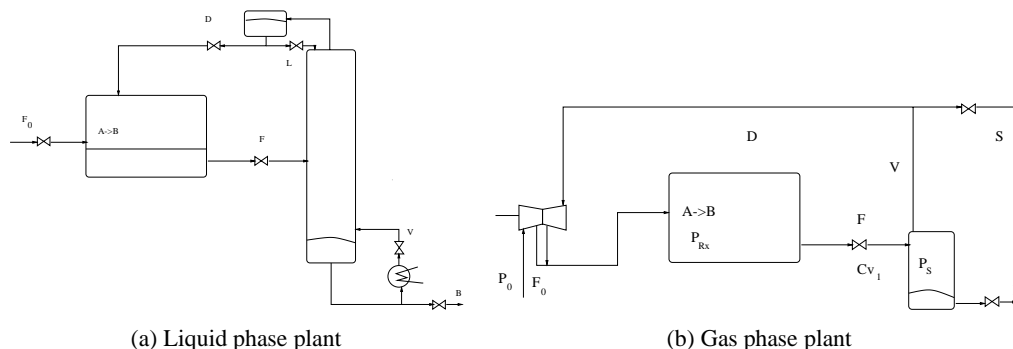(a) Liquid phase plant   (b) Gas phase plant

Figure 1: A gas phase and a liquid phase reactor with separator and recycle.

From the literature it is a well known that recycle changes the behavior of the plant. Gilliland *et al.* (1964) studied a plant similar to 1(a). They showed how the time constant for the whole plant was larger than was expected from the time constants of the individual units, and high gain in the recycle flow for changes in the feed flow.

Verykios and Luyben (1978) studies a similar plant with two reactants and inert. They claim that the process time constant will decrease with increasing recycle and that the dampening depended on distillation dynamics.

Papadourakis *et al.* (1987) used the reactor, separator and recycle plant to demonstrate that the relative gain array (RGA) for the individual units are different than the RGA for the whole plant. The difference is due to the fact that the mass recycle will change the interactions in the plant.

Wolff *et al.* (1992) looked on the effect of recycle on the controllability for a process that included inert. The main focus was on the control of inert composition, with either the recycle, purge or the split fraction between purge and recycle.

Price and Georgakis (1993) looks at a large number of control structures for the reactor separator and recycle plant. From the a large number of simulations with different control structures they try to make some general guide lines. They used a integrated absolute error of product composition as ranking criteria.

Luyben and Floudas (1994) look into the interactions between design and control applied to the plant in 1(a). Their idea is to include controllability objectives as part of a multi-objective optimization. Non-inferior sets shows how controllability can be improved as cheap as possible. They claim that process controllability improved as reactor holdup decreases and as the number of trays in the column increases. A claim valid only for non-inferior sets.

Luyben (1993a, 1993b, 1994) has studied several recycle process. In (Luyben 1994) he discusses the snowball effect: the high sensitivity in the recycle flow rate to changes in feed flow rate. As a remedy for the snowball effect he proposes to fix a flow in the recycle loop.

Wu and Yu (1996) follows up the work of Luyben (1994). They show that for a fixed reactor effluent flow, the reactor hold-up has a high gain for feed-rate changes. Their solution is to control reactor composition.

Ng and Stephanopoulos (1998) also discusses control of the plant in figure 1(a). As Wu and Yu (1996) they also notes the large variations in the volume for feed-rate changes in Luyben's control structure. In addition to the conventional control configuration they proposes to use the reactor temperature to control reactor composition.

The gas phase plant has not been studied to the same extent by academic researchers. Hansen (1998) have studied the methanol synthesis loop, he used a very complex model and did not discuss the selection of outputs. (Loe 1994) discusses the industrial practice for control of ammonia plants, and concludes that it might not be needed to control the pressure.

The work of Fisher *et al.* (1988) must be mentioned here. On page 613 there is some heuristics for plant control. One such heuristics is: "keep the gas recycle flow at its maximum value". They advocates to keep the recycle flow at its maximum value in order to increase the yield. They used the HDA as an example.

Much of the above mentioned works does not discuss the selection of outputs. Two possible is the work of Wu and Yu (1996) and Luyben (1994), however they did not consider the economic impact of their selections. This work will focus on the selection of outputs. To address the issue in a systematic manner we will use the concept of self-optimizing control (Skogestad *et al.* 1999). This involves searching for the variables, which when kept constant give the minimum operating cost. Our conclusions are supported by the use of simple models (isothermal, first order kinetics, etc.). Matlab files of the models are available from the authors.

Outline

First we will look on a simplified plant, where we will look at steady-state relationship. After that we will discuss the selection of outputs for the liquid phase system, and for the gas phase system. The tool is the concept of self-optimizing control, (Skogestad *et al.* 1999). For the liquid phase system we have performed a controllability analysis. Then in conclusion we will elaborate on the difference between gas and liquid system, and how it can give different self-optimizing control structure.

Limitations

In this work we only use simple models, which does not include any energy balances. This is done since normally the temperature in the reactor is given from kinetic considerations.

2 Simplified analysis

2.1 Some steady-state relations, no inert

If we assume first order reaction $A \rightarrow B$, perfect separation and no inert we get the following steady-state equations

$$kM_r z = z_0 F_0 \tag{1}$$

$$B = F_0 \tag{2}$$

$$\frac{B}{F} = (1 - z) \tag{3}$$

$$\frac{D}{F} = z \tag{4}$$

Where k is constant (since we assume constant temperature), M_r is the reactor molar hold-up, z_0 is the composition of A in the feed F_0 (the rest is B), B is the product flow, F is the reactor outlet flow-rate feed to the separator, z is the reactor concentration of reactant A and D is the recycle flow.

The conversion of component A to product is given by $k(T)M_r z$. To increase conversion one has three options:

1. Increase the temperature which increases the reaction constant k.

2. Increase the fraction of A in the reactor, z. This can be done indirectly by increasing the amount of recycle of A.

3. Increase the reactor holdup M_r.

Since at steady-state with given product specifications the conversion of A in the reactor is given by the feed rate, it follows that only one of the two remaining variables mentioned above can be controlled independently (or more generally, one variables that influences these variables), and we must let the second variable "float" and adjusts itself.

There are 7 unknowns in 4 equations, thus given F_0 and z_0 we have one degree of freedom left, e.g. for M_r

$$D = \frac{(F_0)^2}{kM_r - F_0} \tag{5}$$

This means that there is a lower bound on the reactor holdup, $kM_r > F_0$. If M_r is approaches this lower bound then D will go to infinity and z will go to one. This equation illustrates nicely that the "snowball effect" is caused by a too low hold-up in the reactor.

Expressing M_r as a function of D and F_0 gives

$$M_r = \frac{D + F_0}{D} \frac{F_0}{k} \tag{6}$$

which means that if D approaches zero then the reactor holdup M_r goes to infinity.

From these two equations we see that if there is a economic penalty for the recycle D then M_r should be as large as possible. Likewise if there is a economic penalty for hold-up M_r then D should be as large as possible.

2.2 The role of inert

If there is inert and purge present, we have the following mass balances

$$kM_r z + \frac{z}{z + z_I} S = z_0 F_0 \tag{7}$$

$$\frac{z_I}{z + z_I} S = (1 - z_0)F_0 \tag{8}$$

$$B + S = F_0 \tag{9}$$

$$\frac{B}{F} = (1 - z - z_I) \tag{10}$$

$$\frac{D + S}{F} = z + z_I \tag{11}$$

Where z_I is the concentration of inert in the reactor, z_0 is the concentration of reactant A in the feed (the rest is inert).

There are 5 equations in 8 unknown. Thus fixing the feed leaves us with two degrees of freedom. From (Wolff *et al.* 1992) we have the purge flow that gives infinite recycle

$$S_{min} = \frac{F_0}{2} + \frac{kM_r}{2}\left(-1 + \sqrt{1 + \frac{2F_0(1 - 2z_0)}{kM_r} + \left(\frac{F_0}{kM_r}\right)^2}\right) \tag{12}$$

This equation shows that there is a lower limit for purge flow, and that as this limit is approached then the recycle flow goes to infinity. Furthermore the minimum purge flow depends on reactor holdup, a large reactor hold-up will give a smaller purge flow.

3 Economic analysis of the liquid phase system

In this section we will look on control of the plant shown in figure 1(a). The plant consists of liquid phase reactor, where A is converted to B, followed by a distillation column, where A and B are separated, unreacted A is recycled back to the reactor. The design parameters and model is from (Wu and Yu 1996).

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3.1 Minimization of operation costs

First we would like to minimize the costs of production when the production rate is given. For this plant the operating cost is assumed to be energy usage in the distillation column, V. We have constraints on the product quality x_B and reactor level M_r:

$$minimize \quad V \tag{13}$$
$$subject\ to \quad M_r \leq 2800 \tag{14}$$
$$x_B \leq .015 \tag{15}$$

Our degrees of freedom at steady state are, as defined in (Skogestad *et al.* 1999), $N_{SS} = N_u - N_I = 5 - 2 = 3$ (the number of manipulative variables minus the number of inventories which needs to be stabilized and has no steady state effect). There are no penalties for increasing reactor hold-up, but a high conversion in the reactor gives less unreacted A to be recycled (smaller V). In addition to M_r, x_B was also on the constraint, and therefore they should be controlled.

This leaves one degree of freedom, and the question is how it should be used. Since the purpose is to minimize V, it could be tempting to set it directly. There are however one major drawback, if it is set to a too low value it will not be possible to satisfy out constraints. To ensure feasibility its set-point has to be higher than its optimum. Due to equation 1, $z_0 F_0 = k M_r z$, we can not control z since z_0, F_0, k and M_r are given. This leaves F, L, D, L/D, L/F, L/V and x_D as some of the possible controlled outputs. In figure 2 the loss in vapor boil-up due to control error (measurement and/or model error will always be present) is shown. Control of x_D, L/F or L/D gives small losses.

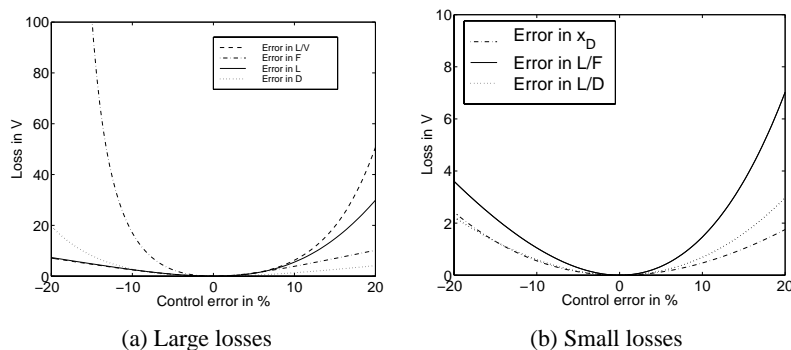(a) Large losses   (b) Small losses

Figure 2: Loss in vapor boil-up, for control errors in selected variables. The nominal values are $L/V = 0.59$, $F = 963$, $L = 713$, $L/F = 0.74$, $L/D = 1.42$ and $D = 503$. For x_D the range is from 0.76 to 0.84 (± 20 impurity)

It is however not enough just to consider the control error, one must also consider the effect of the disturbance on the objective. This effect will depend on which variable that is controlled. In figure 3 we see the loss in boil-up by keeping several variables constant, when F_0 varies with $\pm 20\%$ of it nominal value. And once again, x_D, L/F and L/D seems like good alternatives. But L/F is considerably smaller than x_D and L/D, and we expect L/F to be easier to control than x_D.

The conclusion is that M_r and x_B should be controlled. The ratio L/F should be held constant. But x_D, L/D and L/F has small losses and the final selection can be based on other criteria. Furthermore the Luyben rule, "fix on flow in the recycle loop", can gives very large losses here. In section 4 we have performed a controllability analysis.

3.2 Maximization of production rate

As noted by Rijnsdorp (1991), there are cases where the economic optimum is to produce as much as possible (a sellers market). The purpose of this section is to look into what will limit the production rate, and what consequences this will have for the selection of controlled outputs. The difference from the
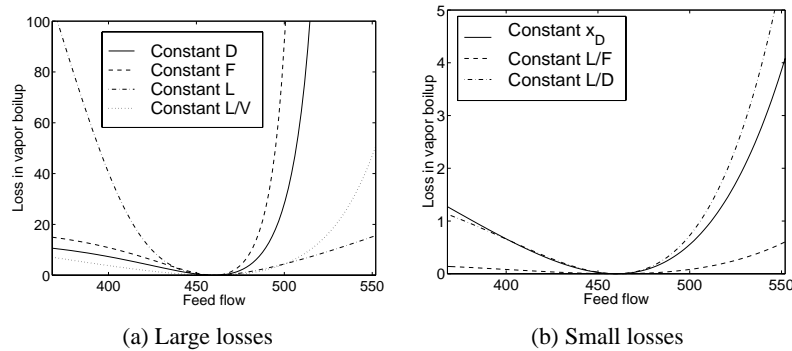
(a) Large losses   (b) Small losses

Figure 3: Loss functions for keeping different variables constant. For all cases x_B and M_r are at the optimum value.

preceding sections is mainly the objective function that is used for optimization

$$minimize \quad -F_0 \tag{16}$$
$$subject\ to \quad M_r \leq 2800 \tag{17}$$
$$V \leq 1400 \tag{18}$$
$$x_B \leq .015 \tag{19}$$

In addition to earlier we have added a constraint on the boil-up in the column, and F_0 is assumed to be a degree of freedom. We will still assume that reactor temperature is not available as a manipulative variable.

What will limit the production rate? Clearly M_r will be at its constraint. x_B will also be on its constraint (if it was purer than the product specification then we could increase production slightly by diluting the product more). The next factor that will influence the production is the composition of A in the reactor (as seen from $r = k(T)M_r z$).

The reactor composition can be affected by using the recycled distillate flow D and the composition in the recycle flow x_D. If we look at the distillation column, by it self, then we have one degrees of freedom left[1]. However for any feed and fixed x_B we know that maximization of V will give the highest possible x_D and D. Thus V should also be at its constraint.
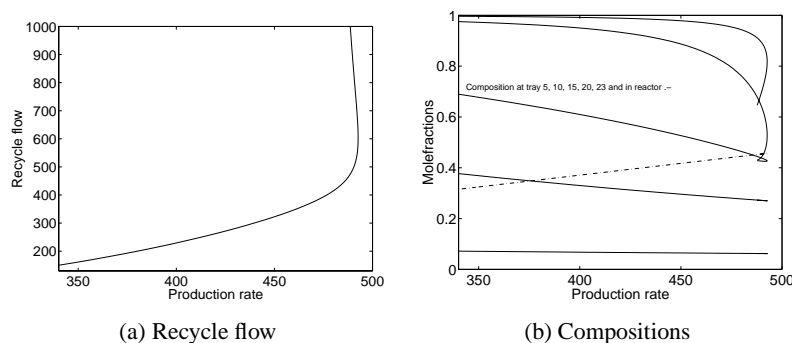

(a) Recycle flow   (b) Compositions

Figure 4: Distillate flow and selected compositions as a function of production rate.

This leaves on degree of freedom. Let us assume that we start with a feed flow of $F_0 = 350kmol/h$, this gives a certain recycle flow D. At low production rates there is almost a linear relationship. But as production is increased the load to the distillation column increases (F and z) and distillate composition x_D decreases. As x_D decrease, more and more D is needed to keep up the production. At $F_0 = 493$ the

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[1]There are four flows D, L, V and B, two levels which must be controlled and one active constraint x_B. Thus the degrees of freedom are $N_{df} = 5 - 2 - 1 = 1$

production rate can not be increased any further. This is shown in figure 4(a). Figure 4(b) shows what happens to some of the compositions in the system. The reactor composition has a maximum attainable value, which will make it unsuitable to use as a controlled variable (the gain will change at the optimum value, and there exists infeasible set-points).

How shall this remaining degree of freedom be used? In figure 5 we have plotted sensitivity for the loss in production rate for keeping different variables at their set-points. (Note that since V is constant, constant L, L/D etc. is the same as constant D.) Neither F_0 or z can be set directly, since we may run into in-feasibility problems (see figure 4). Figure 5 shows that control of D or x_D will give a large sensitivity for control error in these variables to the loss in production rate.
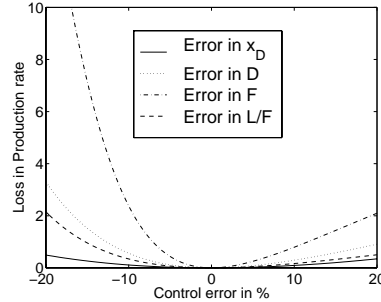

Figure 5: Sensitivity in loss of production for control error in selected variables. The error is $\pm 20\%$ of the nominal value, which is $D = 604 kmol/h$, $F = 1096 kmol/h$ and $L/F = 1.38$. x_D is from 0.85 to 0.78 (± 20 impurity).

However in order to decide which output to control we also need to consider the effects of disturbances. Here we assume that the major disturbance is variations in boil-up. This is a valid assumption since, the boil-up is set indirectly through the heat input. Figures 6[a-b] shows the loss for keeping several variables constant (compared to the optimal value which is a function of the disturbance). And we see that control of L/F gives the smallest loss.
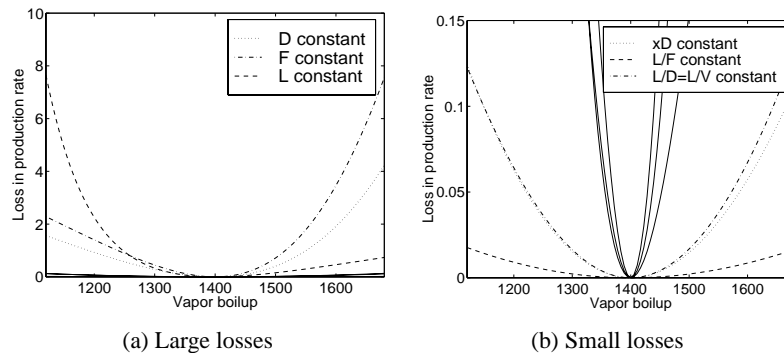

(a) Large losses (b) Small losses

Figure 6: Loss in production rate as a function of vapor boil-up.

To summarize this subsection, we have shown that for maximization of production, M_r, V and x_B should be at its constraints. Control of L/F gives both the smallest loss in production rate for both control errors and disturbance in boil-up.

3.3 The snowball effect

Luyben (1994) states "the use of a conventional control structure resulted in a 100% increase in the recycle flow rate for a 10% increase in the fresh feed flow rate. Such large changes are very undesirable because columns can only tolerate a limited turn-down ratio." His remedy was to fix the reactor effluent flow.

Solving the steady state mass balances for recycle D (Luyben 1994)

$$D = \frac{F_0\left(F_0\left(z_0 - x_B\right) - kM_r x_B\right)}{kM_r x_D - F_0\left(z_0 - x_B\right)} \qquad (20)$$

And we see that as if the reactor holdup is too small then D will approach infinity. Alternatively we can have F and x_D as free variables (Luyben 1994)

$$kM_r = \frac{F F_0\left(z_0 - x_B\right)}{F x_D - F_0\left(x_B - x_D\right)} \qquad (21)$$

Thus for $F - F_0$ and x_B approaching zero we have that the reactor holdup will approach infinity. These effects are by Luyben (1994) referred to as snowball effects.
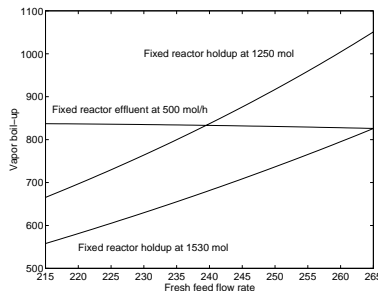

Figure 7: The steady state effect of load changes on boil-up.

If we fix the reactor effluent flow, then there will be very small changes in the vapor boil-up rate. This shown in figure 7 (using the same parameters as in (Luyben 1994). Luyben (1994) had two cases, one with fixed reactor effluent flow and one with fixed reactor hold-up. We have added a case where the reactor hold-up is fixed at the maximum value attained with fixed reactor effluent. The vapor boil-up with fixed reactor effluent, are always higher than with maximum reactor hold-up. There is a steady state cost for letting hold-up float. If there is a lower limit on boil-up this can easily be met (over purification of the product).

4 Controllability analysis of the liquid phase system

In the previous section we have come up with several alternative outputs. The purpose of this section is to look at the controllability of some of these alternatives. We will limit ourself to three cases:

Case 1: Control of x_D, x_B and M_r.

Case 2: Control of L/F, x_B and M_r.

Case 3: Control of x_D, x_B and F.

Case one was by Luyben called the conventional approach. As the section above showed, from an economic point of view, control of x_D is not necessary. Therefor in case two control of x_D is replaced with control of the ratio L/F. In case three we decided to use the Luyben rule, fix one flow in the recycle loop. We want to compare that scheme with case one, therefor we control x_D and let M_r float. (This is not the structure that Luyben proposed for this plant.)

4.1 Poles and zeros in the RHP

The plant has three poles at the origin, and no poles or zeros in the right half plan. These poles are linked to the three levels (M_r, M_D and M_B), which is easily seen from the pole directions. The directions for the pole linked to M_r is

```
y =  -0.9907 xb    u =  -0.4336  F
     -0.0206 xd          0.4621  D
      0.1348 Mr          0       L
      0      Mb          0       V
      0      Md          0       B
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This integrator is different from the other two, it is also observable through the two compositions. This is expected, conversion in the reactor is highly dependent on hold-up. It is feasible to let the reactor float, the unstable pole can be stabilized using other measurements.

4.2   Relative Gain Array and pairing

In figures 20, 21 and 22 in appendix A the relative gain array is plotted as a function of frequency. For the two cases where M_r is included as controlled outputs we see that we have infinite RGA at steady state. This is due to the fact the the integrator associated with reactor level M_r is observable in more than one output (M_r, x_D and x_B) and controllable in more than one input (F and D). According to **Larsson** this will give infinite RGA at steady state. It is however the magnitude of the RGA at crossover frequency that is of most interest.

   If we pair to avoid negative RGA at steady state, and RGA near one at crossover frequency, we get the following pairings:

For case 1: $x_B - V$, $x_D - L$, $M_r - F$, $M_B - B$ and $M_D - D$.

For case 2: $x_B - V$, $M_r - F$, $M_B - B$ and $M_D - D$.

For case 3: $x_B - V$, $x_D - L$, $M_B - B$ and $M_D - D$.

For simplicity we will in the remaining part of the paper assume perfect level control, and we assume that the ratio is implemented as $L = \frac{k}{\tau s + 1} F$. This reduces case one and three to two $2\mathbf{x}2$ plants and case two is reduced to a SISO plant.

4.3   Case 1: Control of reactor level

By closing the level loops the relative gain for the remaining plant is $\Lambda(jw) \approx I$. At first this may seem strange, how is it possible that recycle reduces the interactions? Consider an increase in reflux L; since M_D and M_r are perfectly controlled, it will be followed by an decrease in the feed to the distillation column. This decrease will counter the effect of the increase in L on the change in x_B. The net result is that the gain from L to x_B is small, and thus that we have a triangular system. This effect is confirmed by nonlinear simulations, if the change in L is large ($> 10\%$) then L has an effect on x_B.

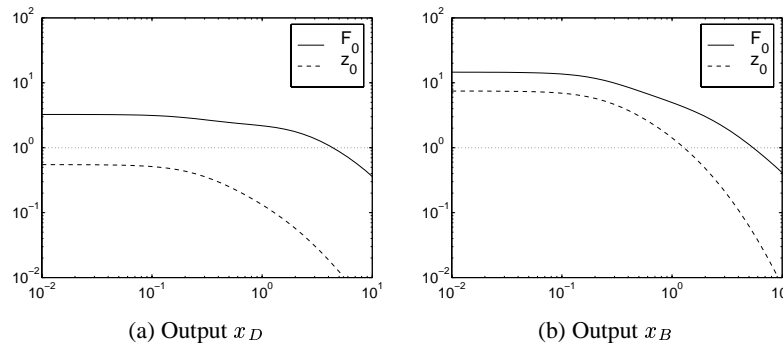(a) Output x_D (b) Output x_B

Figure 8: Closed loop disturbance gain for case one.

   According to the closed loop disturbance gain, (Skogestad and Postlethwaite 1996), a bandwidth of 5 is needed in both control loops, figure 8. w_{180} is well above this bandwidth, a PI-controller will be sufficient.

4.4 Case 2: Ratio control

In figure 9 the disturbance gain is plotted, it shows that we will need a bandwidth of 5, a bandwidth which is achievable with a simple PI-controller.
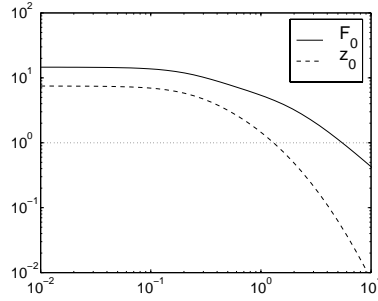

Figure 9: Disturbance gain for case 2.

4.5 Case 3: Floating reactor level

In this plant the reactor level is still floating and we will need to stabilize it. There is a pole in the origin. The pole at the origin, is observable in both outputs and controllable with both inputs. As shown in **Larsson** this gives infinite RGA values at steady state. However the relative gains drops fast and at $w = 10$ we have $\Lambda \approx I$.
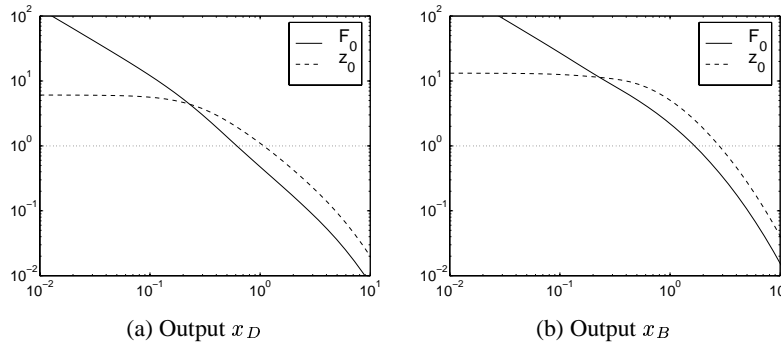

(a) Output x_D (b) Output x_B

Figure 10: Closed loop disturbance gain for case three.

In 10 we have plotted the closed loop disturbance gain array for this plant, and we see that we will need a bandwidth of 1 in the first loop and a bandwidth of 3 in the second loop.

This case does not have significantly better controllability than the other cases. This means that we have not found support for the Luyben rule based on controllability aspects either.

4.6 Observed controller performance

To confirm the the findings above we perform nonlinear simulations, using the following controllers (where k_1 is for x_D, and k or k_2 is for x_B):

- Case 1: $k_1 = 5595\frac{3s+1}{3s}$ $k_2 = -106610\frac{2s+1}{2s}$
- Case 2: $k = -121510\frac{3s+1}{3s}$
- Case 3: $k_1 = 5096\frac{3s+1}{3s}$ $k_2 = -28736\frac{s+1}{s}$

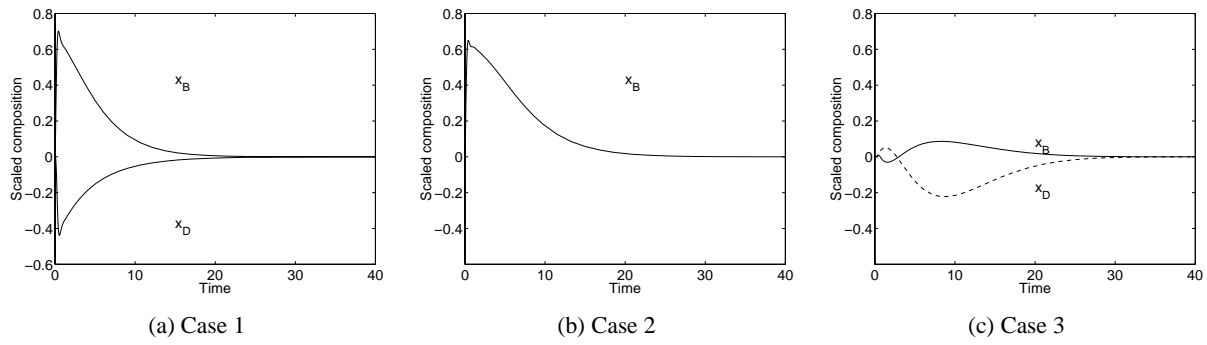The response to a step of 20 % increase in the feed rate, is shown in 11.

| (a) Case 1 | (b) Case 2 | (c) Case 3 |

Figure 11: Closed loop response to a step in feed rate. The mole fractions are scaled.

5 Economic analysis of the gas phase system

In this section we will study the gas phase system in figure 1(b). The plant is not much different than the liquid phase plant: the distillation column is replaced with a separator and two compressors is added. For gas phase systems, inert and purge has an important effect on the operation, and are included in the model. The main assumptions are:

- Sharp split in the separator reactant A and inert is separated from B.
- Ideal gas law.
- The reactor effluent flow to the separator is driven by the pressure difference.
- The compressor work is modeled as isothermal ideal gas compression.

We have assumed that both feed flow rate F_0 and up-stream pressure P_0 are independent variables (disturbances). This is not really possible, since an changes in F_0 must be followed by a change in pressure P_0 (the pressure P_0 represents some local up-steam gas hold-up). So implicitly we have assumed that there is a control system in place (i.e. the main compressor is adjusted to keep P_0 constant).

5.1 Optimization

The goal of operation is to maximize the values of the product stream diminished by the value of feed streams and utility costs. For the liquid phase plant there was only one feed and only one exit stream, hence during the optimization we need only to minimize the cost of utility. However for the gas phase system we have two product flows with different price. The objective function is then

$$J = \$_B B + \$_S S - \$_{F_0} F_0 - Utility \tag{22}$$

Using the total mass balance, $S = F_0 - B$ gives,

$$J = (\$_B - \$_S)B - (\$_{F_0} - \$_S)F_0 - Utility \tag{23}$$

F_0 is given, and the utility is mainly compressor work:

$$J = B - w_r (W_1 + W_2) \tag{24}$$

Where w_r is the relative cost of compressor work compared to $\$_B - \$_S$. W_1 and W_2 is the main and recycle compressor work. The main compressor work is given by

$$W_1 = F_0 RT \ln \frac{P_{Rx}}{P_0} \tag{25}$$

This increases with P_{Rx}. So, unlike the liquid phase system, there is a penalty for increasing the hold-up in the reactor. If the upstream pressure is higher than P_{Rx} then there would be no need for the main compressor and this term will vanish. The recycle compressor work is given by

$$W_2 \quad = \quad DRT \ln \frac{P_{Rx}}{P_S} = DRT \ln \frac{P_{Rx}}{P_{Rx} - \Delta P} \tag{26}$$

$$\approx \quad \frac{RTD\Delta P}{P_{Rx}} \approx \frac{(z + z_I)cRTF^2}{P_{Rx}} \tag{27}$$

The first approximation is valid when pressure drop ΔP in the loop is small compared to P_{Rx}. The second approximation relates the pressure drop to the flow ($\Delta P \approx cF$). Here we see that the work increases approximately with F^2. In addition, to reduce the factor c all flow restrictions should be as small as possible, i.e. all valves in the loop should fully opened. A large pressure will decrease this term, directly through P_{Rx}, and also since a high pressure gives a high conversion which reduces z. This term is often negligible compared to the main compressors work. Therefor the two compressors are often mounted on a common shaft, which will be the case in this study (see section 6.3.1).

The last term in 24 represents the valuable product. Hence it should be as large as possible. Production rate can be increased by increasing the reactor pressure and there by decreasing purge. This gives a increase in compressor work. If the relative cost of compression is low the optimum will be constrained, either maximum pressure or maximum compressor load. This is not always the case, as we approach full conversion the cost of compressor work can become significantly.

5.2 Degrees of freedom

We have assumed that feed rate is given, this implies that the main compressor work is given and therefore also the recycle compressor work. Since any valves in the loop should be fully open, we only have two manipulative variables. One of these B has to be used for controlling the liquid level in the separator. Which leaves us with one degree of freedom, here chosen as purge flow S.

5.3 Optimization

Figure 12 shows the elements involved in the objective function. As purge flow is reduced, we produce more valuable product. But the pressure increases and, the compressor work increases. At full conversion, the reactor pressure and compressor work would be infinite.

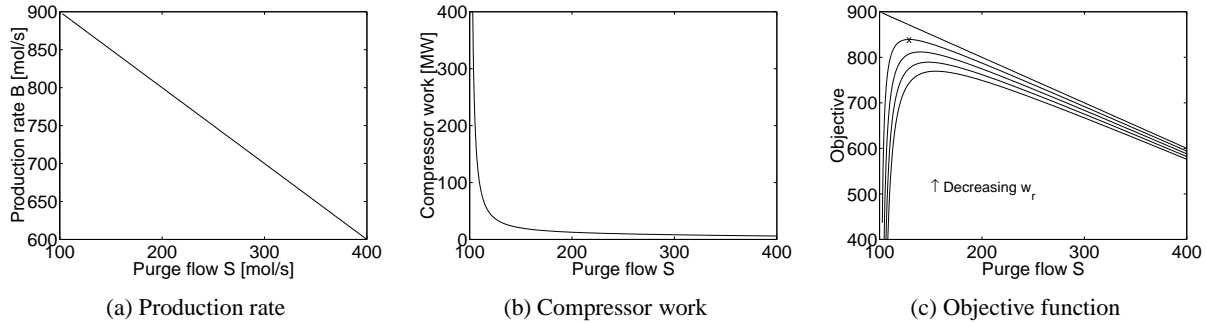(a) Production rate   (b) Compressor work   (c) Objective function

Figure 12: Terms in the objective function and the objective function, with different relative weights, as a function of purge flow.

In figure 12(c), the objective has been plotted for several relative costs. It shows how the optimum would move towards higher conversion if the relative weight is reduced w_r. If w_r is low enough the optimum will be constrained, either by maximum pressure or by compressor load. We will study a unconstrained case, with a relative weight of $1 \ mol/MJ$, marked with an x in figure 12(c).

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5.4 Self-optimizing control

Our candidates for self-optimizing control are: purge flow S, recycle flow R, purge flow fraction S/R, inert composition in recycle z_I and reactor pressure P_{Rx}.

5.4.1 Control error

Table 1 shows the loss for the expected control error.

Variable	Range		Max. loss
S	103 - 155	mol/s	330
R	2230 - 3345	mol/s	1.7
S/R	0.037 - 0.055		1.2
z_I	0.676 - 0.876		18.2
P_{Rx}	189 - 231	bar	1.0

Table 1: The worst loss within the range.

There are two variables which clearly are very sensitive to control error, S and z_I. If we look at figure 12, we see that a small error in purge flow could lead to a large drop in the objective. When we approach the lower limit on purge flow, the inert composition z_I approaches one. Thus there are only a small gain between S and z_I, which implies that an error in z_I will be present in S. P_{Rx}, will however approach infinity as S is reduced. There will be a high gain between S and P_{Rx}, which implies that a large error in P_{Rx} will become a small error in S. Similar arguments applies for R and S/R.

5.4.2 Loss due to disturbance

We will only consider the effect of holding P_{Rx}, R and R/S when feed rate changes. As we can see from figure 13, there are only small differences, and the final choice should be based on other considerations.
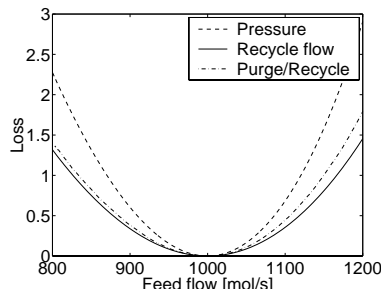

Figure 13: The loss from optimality by holding selected variables constant.

6 The methanol synthesis loop

The methanol synthesis loop is one part of a methanol plant. The feed comes from the reformer section, where synthesis gas is produced from natural gas. The product flow is sent to the distillation columns. There exist several good references which describes the methanol synthesis, (Olsvik *et al.* 1997), (Lee 1990), (Skrzypek *et al.* 1994) and several more. Only a brief overview of the process is given here.

Process layout

The feed (which consists of CO, CO_2, H_2O and H_2) is compressed to the reaction pressure, which is approximately 80 bar. Next it is preheated in the feed/effluent heat exchanger. In the reactor the catalyst

is packed in vertical tubes, which are surrounded by boiling water. The temperature is 250 ^oC. After the reaction the product is cooled, and the product are separated in flash drums. The overhead gas is divided into purge and recycle.
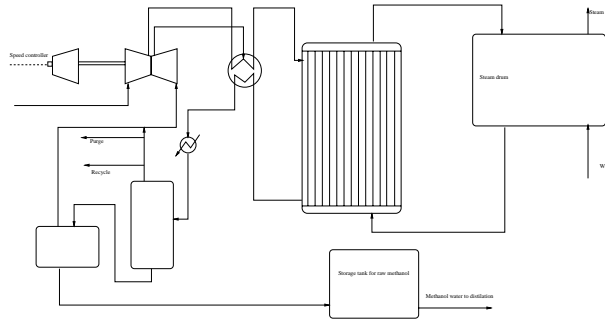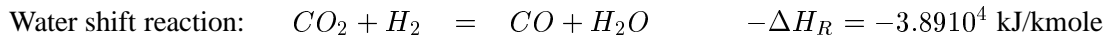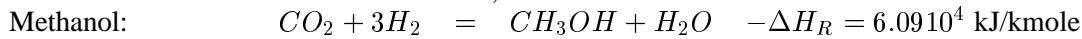

Figure 14: The methanol synthesis loop.

Reactions

Two reactions are considered in the reactor, these are:

Methanol:	$CO_2 + 3H_2$	$=$	$CH_3OH + H_2O$	$-\Delta H_R = 6.09 10^4$ kJ/kmole
Water shift reaction:	$CO_2 + H_2$	$=$	$CO + H_2O$	$-\Delta H_R = -3.89 10^4$ kJ/kmole

Commercial methanol catalyst are highly selective and at the operating conditions (high temperature and H_2 in excess) the production of side product are negligible.

Modeling

The models are simple, but the most important features are captured. The reactor is modeled as a series of isothermal CSTR with kinetic model from (Vanden Bussche and Froment 1996).

One important factor for the optimal operation of the synthesis loop is that the flash drum does separate product and reactant. This is achieved by cooling the reactor effluent sufficiently. The solubility of CO_2 in the liquid phase is in the order of 1%. We assume perfect separation of liquid and gas in the flash drum.

Since we have assumed isothermal reactor and perfect split in the separator we can ignore the heat exchangers.

The compressor work is modeled as polytropic compression of ideal gas. Since they are mounted on a common shaft, the work of the recycle and main compressor is related by $w_{recycle} = k w_{main}$ where w is the molar work and k is a constant. The reactor effluent flow is given by $C_V \Delta P$, where C_V is chosen to give reasonable pressure drop in the loop.

6.1 Degrees of freedom analysis

If all temperatures are controlled and that the feed is given, then purge flow and product flow are the manipulative variables. (The recycle flow rate is given by the compressor work and the pressure drop in the loop. The optimum the pressure drop should be as small as possible.) There is one inventory without steady state effect which must be controlled, liquid hold-ups in the flash drum. This gives us $N_{df} = 2 - 1 = 1$ degrees of freedom for optimization.

6.2 The objective function

The key element for selection of controlled variables is the economic objective. In this case there are one important effect which will not be captured with steady state economics. Namely the activity versus the deactivation rate of the catalyst. This subject has been studied by Løvik *et al.* (1998). They showed

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that there is a optimal temperature profile as a function of time, but they did not discuss different ways of implementing their optimal policy. Since deactivation mainly depends on reactor temperature, we will assume that the reactor temperature is given. Therefor steady state economics will be used on the remaining degrees of freedom.

We have also done one more assumption: that the synthesis loop is decomposed from the rest of the plant. Both the feed and the product flow to the synthesis loop are internal flows in a larger plant. There are a small recycle flow from the synthesis loop to the reformer section. We do believe that the two-way interactions are small enough to allow for a vertical decomposition of the process for selection of the controlled variables.

The objective function is

$$J_1 = \$_B B + \$_S S + \$_P P - \$_{F_0} F_0 - Utility \tag{28}$$

Where B is product flow, S is purge steam, P is steam, F_0 is the feed, and $Utility$ is the cost of utility (compressor work).

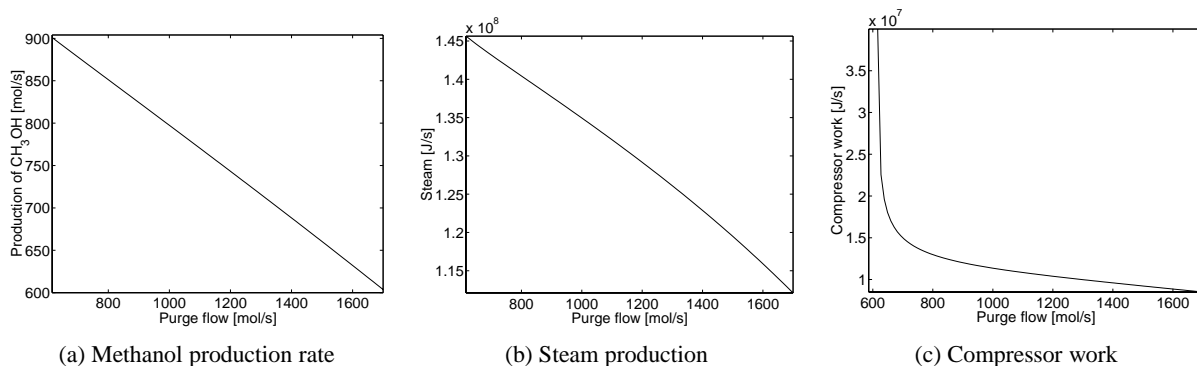(a) Methanol production rate   (b) Steam production   (c) Compressor work

Figure 15: Terms in the objective function as a function of purge flow.

How are the different terms in the objective function influenced by our degree of freedom? Figure 15 shows the production of methanol, steam, and consumed work against the purge flowrate. As the purge flow approaches the minimum purge flow $S_{min} = 607 \ mol/s$ (full conversion, see appendix B), then the recycle flow and reactor pressure goes towards infinity. This can be seen from figure 15(c), where compression work approaches infinity. The remaining degree of freedom will have an optimal value.

As shown in appendix B, there is a linear relationship between purge and product flow, and figure 15(b) shows that the produced amount of steam is almost a linear function of purge. Therefor we choose to use a simplified objective

$$J = B - w_r W \tag{29}$$

Where B is the production of methanol, W is the compressor work and w_r is the relative cost.[2].Figure 16 shows the dependency of the objective function for different values of w_r. For all the cases of $w_r \neq 0$, there is a steep fall only on one side of the optimum. The smaller the value of w_r the closer the optimum is to the steep fall, which is easily explained by figure 15. We will study an unconstrained optimum ($w_r = 1e^{-5}$).

6.3 Self-optimizing control

The remaining degree of freedom should be chosen such that the economic performance shows small sensitivity to disturbances and control errors. Some alternatives are: Reactor pressure, composition of

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[2]From J_1 one can derive an exact definition of w_r. However the reason for introducing the simplified objective is to have fewer parameters in the objective function.
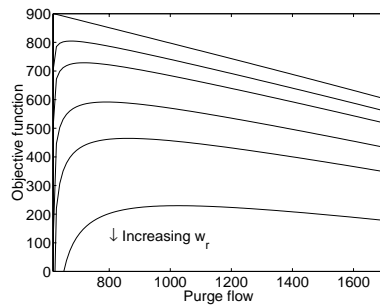
Figure 16: The objective function for different values of the relative cost ranging from 0 to $3e^{-5}$.

hydrogen in the recycle loop, composition of inert in the recycle loop, constant purge flow, constant recycle flow or constant purge fraction.

Figure 17 shows the loss function for three disturbances. For feed rate variations constant purge flow (S) gives a very large loss, constant reactor pressure (P_r), recycle flow (R) or recycle ratio (S/R) gives a moderate loss, while control of inert or hydrogen composition in the recycle flow gives a very small loss. For variation in inert compositions in the feed only constant inert composition in the recycle flow gives a large loss. And from the last figure 17(c) we see that P_r, R and z_{H_2} has the smallest loss for variations in stoichiometric number.

(a) Feed rate

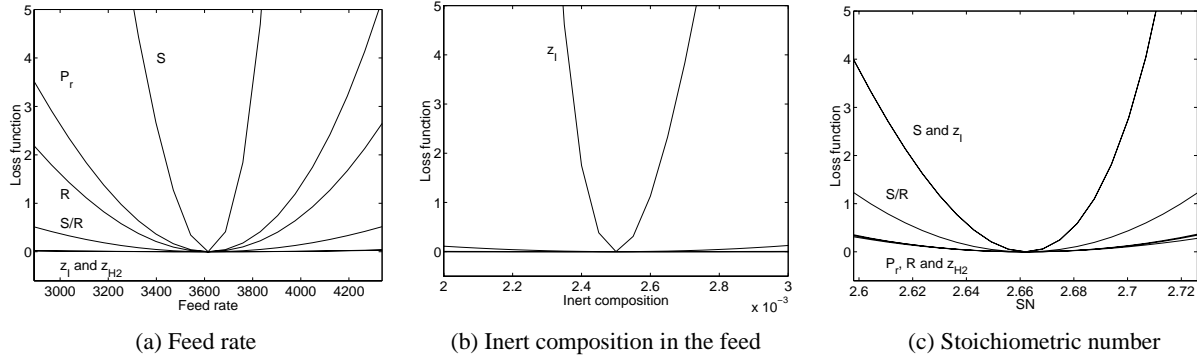(b) Inert composition in the feed

(c) Stoichiometric number

Figure 17: The loss for keeping variables constant under disturbances. S is constant purge flow, P_r is constant reactor pressure, R is constant recycle flow, S/R is constant purge recycle ratio, z_I is constant inert composition and z_{H_2} is composition of hydrogen in the recycle flow.

From figure 18 we can see that purge flow, inert composition and hydrogen composition in recycle has largest sensitivity to control errors. Control of the purge/recycle ratio, recycle and pressure gives the smallest. In summary one should control either S/R, R or P_{Rx}. The results for this case conforms to the results for the simple gas phase system.

6.3.1 The common shaft

To save investment costs the compressors are mounted on the same shaft. This means that we for operation looses one degree of freedom. In 19 shows how we could have improved the objective if recycle work was an independent variable. The back-off from the operating optimum is due to the difference between design optimum and operating optimum.

The difference between the current point, and the operational optimum is so small that it will probably not be justifiable to have a separate shaft for the recycle compressor. Since the recycle/main compressor work is not on the operational optimum, there are no point in calculation of the loss. (However variations in disturbances should be considered during the design.)
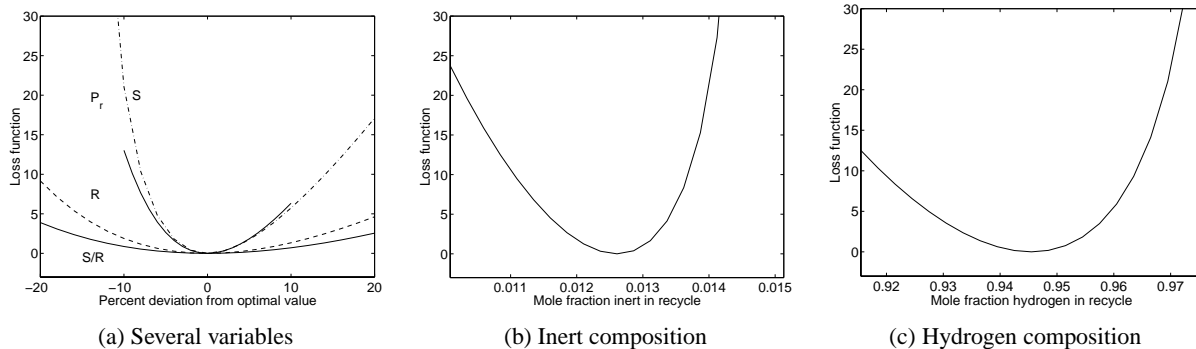
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|(a) Several variables|(b) Inert composition|(c) Hydrogen composition|

Figure 18: Losses due to control error in controlled variables. S is constant purge flow, P_r is constant reactor pressure, R is constant recycle flow and S/R is constant purge recycle ratio.
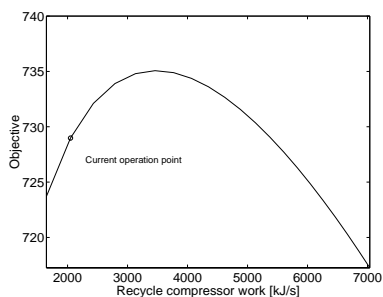

Figure 19: The objective as a function of recycle compressor work.

However if there there where two different shafts, then we could increase the recycle compressor work without increasing the size of the compressor. But as indicated by figure 19 the optimum would most likely be at a constraint for the compressor. This argument is due to Fisher *et al.* (1988).

7 Discussion and conclusion

In this paper we have looked on the selection of variables for recycle processes. We looked at two main cases, liquid phase system and gas phase system. There are fundamental differences in the economic operation of these systems which must be taken into considirations when designing a control system. Our focus has been on the selection of controlled variables.

For the liquid phase system we studied two different objectives: minimization of the operating cost, which was the boil-up, and maximization of production rate, constrained by boil-up. In both cases there was no cost associated with increasing hold-up, therefor its optimum was to at its constraint. Floating reactor hold-up would mean a steady state economic loss. In both cases control of x_D or L/F was good candidates for self-optimizing control.

In contrast to the generic rule for recycle system (Luyben 1993c): "one flow rate somewhere in the recycle loop should be flow controlled.", keeping a flow in recycle loop, can give a very steep loss function. Which makes optimization difficult. We did not find any indications that the Luyben rule would give a plant with better controllability. (We are not saying the opposite, which would be never to fix a flow in recycle.)

For the gas phase system the situation is different, there are a cost ascosiated with reactor hold-up (pressure). The optimum are unconstrained in this variable. Fixing recycle rate, purge fraction, or reactor pressure had all good self-optimizing properties. This is linked to the behaviour of these variables as conversion increases. As expected purge flow is a bad alternative as a controlled variable. More unexpectedly, inert composition in the recycle turned out to bad self-optimizing properties. This is also

explainable by the behaviour of this variable when conversion is increased. The results for the gas phase reactor carries well over to the methanol case study.

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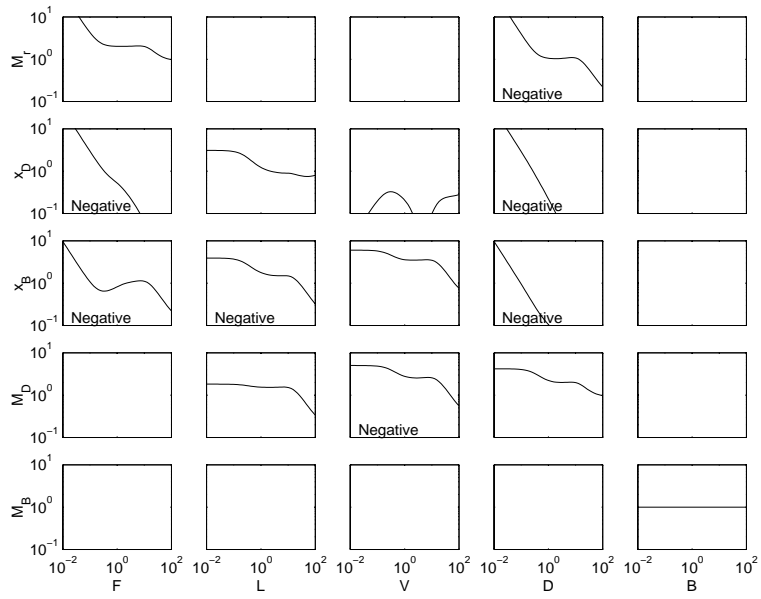
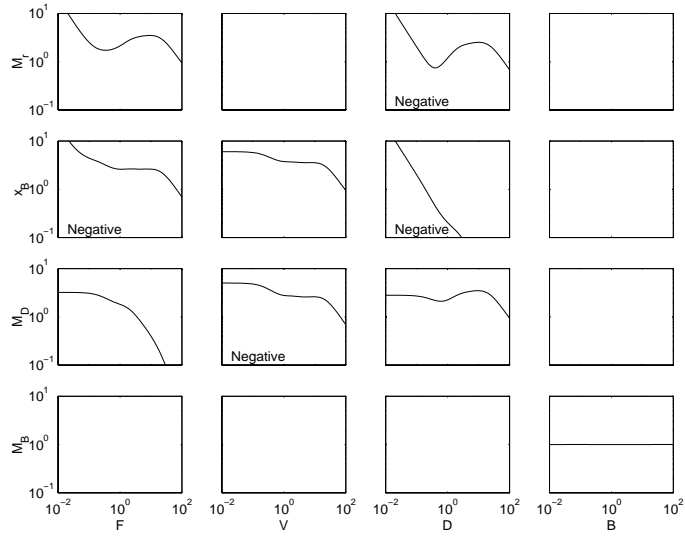Figure 20: Relative gain array for case 1.

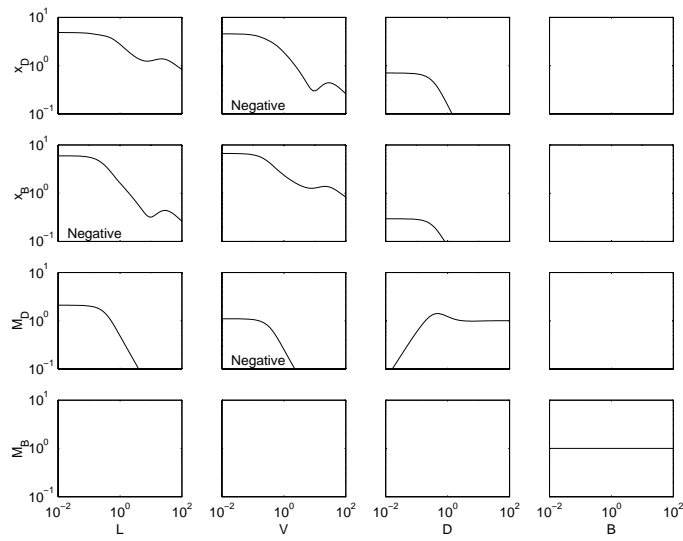Figure 21: Relative gain array for case 2.

Figure 22: Relative gain array for case 2.

A RGA

B Some simple relations

In order to understand some of the most basics facts of the methanol synthesis, we can derive some simple equations based on atomic balances. All results in this appendix are trivial and well known, but still important.

Inert balance:
$$Sy_{CH_4} = F_0 z_{CH_4}$$

Carbon balance:
$$S(y_{CO_2} + y_{CO}) + Bx_{CH_3OH} = F_0(z_{CO_2} + z_{CO})$$

Oxygen balance:
$$S(2y_{CO_2} + y_{CO}) + B(x_{CH_3OH} + x_{H_2O}) = F_0(2z_{CO_2} + z_{CO} + z_{H_2O})$$

Hydrogen balance:
$$Sy_{H_2} + B(2x_{CH_3OH} + x_{H_2O}) = F_0(z_{H_2} + z_{H_2O})$$

Where we have assumed that there is no methanol in the feed, and that the are no gases in the product and no liquid in the the purge flow. In addition the mole fraction must sum to one in all the flows. For a given feed we then have six equations in eight variables, thus there are 2 degrees of freedom. (These equations does not say anything of the internal flows and pressures.)

For a given feed, with surplus of H_2 will have maximum production if all CO and CO_2 is converted to methanol. That is in the limiting case $y_{CO} = y_{CO2} = 0$. Then the production of methanol would be

$$x_{CH_3OH}B = F_0(z_{CO_2} + z_{CO}) \tag{30}$$

The minimum amount of hydrogen in the purge flow

$$Sy_{H_2} = F_0(z_{CO_2} + z_{CO})(SN - 2) \tag{31}$$

Where the stoichiometric number is defined as $SN = \frac{[H_2]-[CO_2]}{[CO]+[CO_2]}$. Which together with the amount of inert gives the minimum purge flow

$$S = F_0(z_{CH_4} + (z_{CO_2} + z_{CO})(SN - 2)) \tag{32}$$

If $SN = 2$ then we will lose no hydrogen in the purge for full conversion.

What these equation tells us is the obvious, for a given feed there is a upper limit for produced methanol which is given by the amount of carbon in the feed. And there is a lower limit for purge gas, which is given by the amount of inert and surplus of hydrogen in the feed.

The remaining part of this section will look at the relation between purge and product when the feed is given. The purge flow is

$$S = S(y_{CO} + y_{CO_2} + y_{H_2} + y_{CH_4}) \tag{33}$$

Where we can insert the atomic balances:

$$S = F_0 - B(3x_{CH_3OH} + x_{H_2O}) \tag{34}$$

Which also can be derived from a total mole balance.

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