Design and Control of Recycle Systems
by Non-linear Analysis

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Abstract

Design and control of recycle systems must be integrated at an early design stage. By placing together the pieces of the design puzzle we developed a novel methodology that allows the screening and selection of feasible integrated designs at an early stage.
The generic structure is the Reactor-Separator-Recycle (R-S-R) system. Multiple steady states are possible and a minimum reactor volume is required. The behaviour depends on the reactor size and plantwide control structure. Larger reactors behave better than smaller ones. A clear distinction is made between *self-regulation* and *regulation-by-feedback* control structures. For some complex reactions, the simple self-regulation feed policy is effective and ensures the desired selectivity pattern. This study integrates the results into a new reliable methodology for design and control of recycle systems.

Introduction

The integrated design and control of recycle systems in the frame of non-linear analysis was largely ignored by now both by designers and control engineers. A review of the literature on integration of design and control shows that applications of non-linear tools to design of recycle systems is a recent subject (Seferlis & Georgiadis, 2004).

Designing simultaneously the reactor and separators is difficult in practice due to the complexity of each unit. The functional analysis should establish if the system's architecture is feasible and operation stable or not. By placing together the design and control aspects in a systemic frame we have developed a novel design methodology based on non-linear analysis. This allows the quick screening and selection of feasible integrated designs at an early conceptual stage. The methodology was successfully applied to several case studies involving complex stoichiometry (Kiss et al., 2005).

This article begins with an overview of non-linear effects of importance for recycle systems. Next, we show that a minimum reactor volume is required. In addition, the feedback effect of recycles can generate state multiplicity. For safe operation the reactor should be sufficiently large. The major plantwide control structures are analysed and the main features are discussed. The control structure determines the type of behaviour exhibited by the system. Contrary to the stand-alone analysis, in recycle systems different selectivity can be achieved only by adequate fresh feed policy. Moreover, the selectivity does not exhibit a maximum against conversion or residence time.
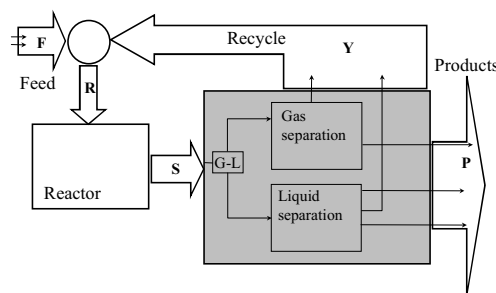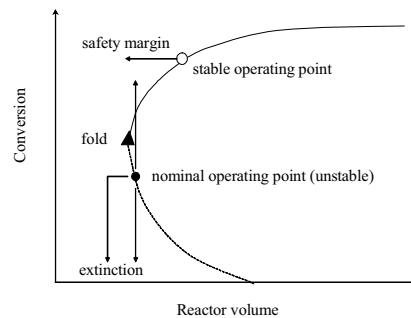
Figure 1. Generic flowsheet of R-S-R   Figure 2. Non-linear phenomena in R-S-R.

Non-Linear Behaviour of Recycle Systems

The non-linear behaviour of recycle systems is relevant for integrating design and plantwide control at an early stage, when the recycle structure of the flowsheet is established (Fig. 1). In recycle systems, important non-linear effects are possible: high sensitivity, unfeasibility and state multiplicity. These phenomena may appear in case of self-regulation control structures. However, they are not a problem if the reactor is sufficiently large. In addition, they can be avoided if the reactor-inlet flow rate is fixed but in this case the production rate and selectivity are more difficult to change.

High-sensitivity or the so-called *snowball effect* consists of getting a non-linear large amplitude response, to a relatively small change in parameters. Avoiding the snowball effect by proper design of the whole system is a better solution than oversizing the separators. Luyben et al. (1999) showed that snowball effects are responsible for difficulties in process control of systems with recycle. They also developed a *constant-recycle* rule that have a great conceptual value in building effective plantwide control strategies. As shown by Bildea & Dimian (2003) the control problems created by snowball effects can be simply treated by proper reactor design. Snowball effects are caused by small reactors that had to be compensated by large recycles. When the reactor is sufficiently large the snowball effect disappears and classical control structures apply.

Multiple steady states can occur in recycle systems, due to the solely feedback effect of material recycles (Fig. 2). In addition, regions where no feasible states exist are possible. The operation in a stable point is obviously recommended. However, designing the system in a stable region but close to the turning point could be dangerous due to high sensitivity. The bifurcation diagrams are powerful tools in selecting the nominal design that guarantees stable and safe operation.

Minimum Reactor Volume

In contrast to the stand-alone approach, the volume of a reactor placed in a recycle system must exceed a critical value to ensure a stable operation. The demonstration was performed initially for simple reactions (Bildea et al., 2000), and later extended to more complex reactions including polymerisation systems by Kiss et al. (2003).

The plant Damköhler number $Da = k_1 \cdot (V/F_0) \cdot C_{A,0}$ conveniently groups three variables characterising the material production: flow-rate of reference reactant, reactor volume and reaction rate – by means of kinetic constant.
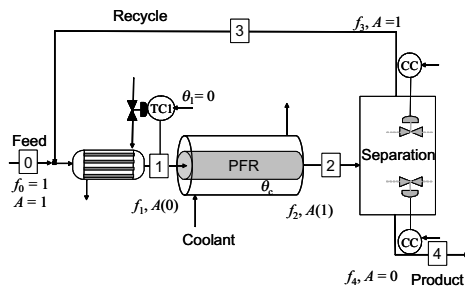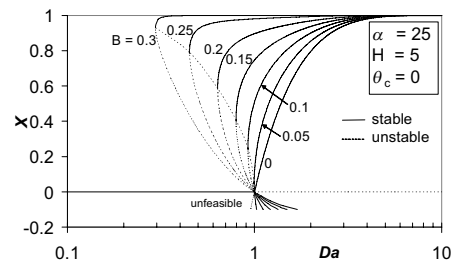
Figure 3. PFR - Recycle system.

Figure 4. Da-X bifurcation diagram.

We consider as example the first-order reaction $A{\to}P$ carried out in a PFR recycle system (Fig. 3). Fig. 4 presents the bifurcation diagram for different values of the adiabatic temperature rise ($B{\geq}0$), using conversion as dependent variable. This diagram can be traced using Matlab or other software for continuation and bifurcation analysis. At the transcritical bifurcation point $Da^{T}=1$, two different manifolds cross each other in the combined space of state variables and parameters. The entire bifurcation diagram contains one turning point that enters the feasible region at a boundary limit singularity. An exchange of stability takes place at the transcritical bifurcation. The instability of the low-conversion state can be proven by steady state analysis (Bildea, 2004). The limiting value $Da=1$ is independent of the reactor type. Such feasibility constraint is typical to recycle systems and does not appear in case of stand-alone reactors (Kiss et al., 2003).

Figure 5 shows that a minimum reactor volume exists as well for the reversible reaction $A{\leftrightarrow}B$ taking place in an isothermal recycle system with incomplete separation (Fig. 3). The transcritical value is given by: $Da^{T} = \beta{\cdot}(1-z_{A,4}) \ / \ (z_{A,3}+\beta{\cdot}z_{A,3}-1)$. For complete separation ($z_{A,3}=1$, $z_{A,4}=0$) the limiting value is again $Da^{T}=1$. The comparison between CSTR- and PFR-Separator-Recycle systems shows that reactor volumes ratio increases for higher conversions, thus the same conversion can be achieved in a smaller PFR.

State Multiplicity

Sizing reactors in recycle systems implies considering the state multiplicity. Multiple states can occur exclusively due to the mass feedback (Bildea et al., 2000). We consider the second order reaction $A+B{\to}P$, taking place in an isothermal CSTR recycle system. When reactants are fully recycled only one feed may be set on flow control ($f_{A,0}=1$).The second reactant ($f_{B,0}$) must be added in a way that respects the overall material balance.
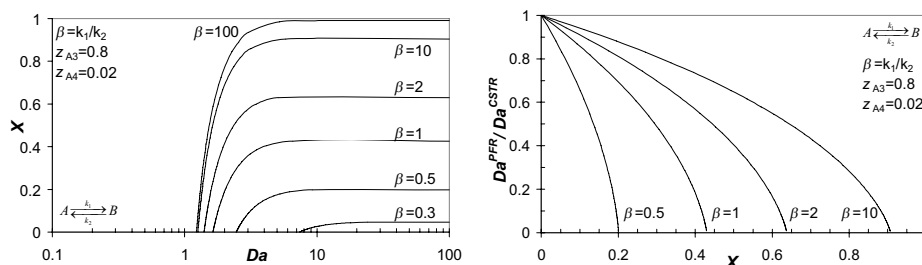

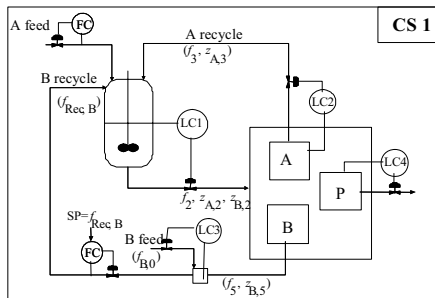*Figure 5. Da-X bifurcation diagram. Reactor size (PFR/CSTR ratio) vs. conversion.*
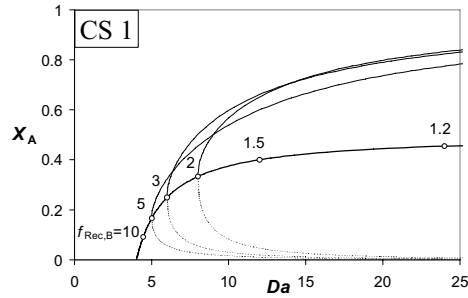
Figure 6. Regulation-by-feedback control.

Figure 7. State multiplicity in R-S-R.

Two control structures are possible: **CS1** - fresh A on flow control, fresh B in recycle loop, fixed recycle of B (Fig. 6); and **CS2** - fresh A on flow control, fresh B on level control, fixed reactor exit flow rate. For CS1, the conversion of the key component X_A is shown in Fig. 7. In case of CS2 the behaviour is similar to CS1. For given values of the fixed flow rate ($f_{Rec,B}$ or f_2) and separation performance ($z_{A,3}$ and $z_{B,5}$), two feasible steady states exist when Da exceeds the critical value corresponding to the turning point of the Da-X_A diagram. The lower state is unstable and has an unusual behaviour: a larger reactor gives lower conversion. The instability can be proved based on steady state considerations, showing that the analogue of CSTR's slope condition is not fulfilled (Bildea et al., 2000). A recycle system in which the reactor is designed near Da^{cr} can suffer from serious operability problems due to high-sensitivity around the fold and proximity of region with no feasible states ($Da < Da^{cr}$).

Selectivity in Recycle Systems

Selectivity is a key topic for the design of reactors in recycles, but classical textbooks discuss it exclusively from a stand-alone viewpoint. Reactor type, CSTR or PFR, conversion level and mixing method of reactant are means to manipulate selectivity. Typically, low conversion and PFR's are recommended for good selectivity. In R-S-R, the recycle policy and plantwide control of feeding the reactants play the determinant role in achieving desired selectivity, and not the reactor type or conversion.

Let us consider the consecutive/parallel reactions: $A+B \rightarrow P$, $A+P \rightarrow R$, where reactants have adjacent volatilities and they are recycled together (Fig. 8).
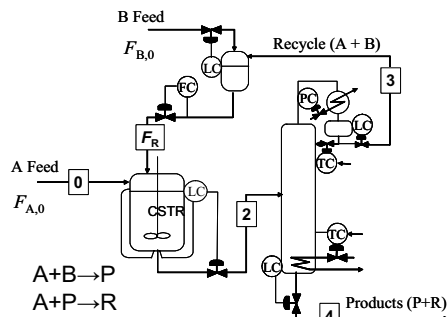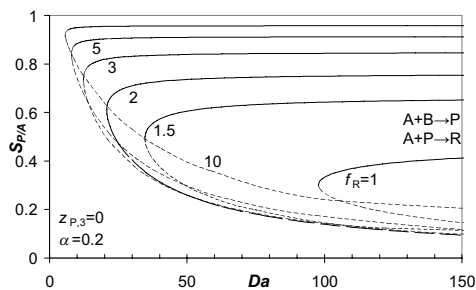

Figure 8. Self – regulation structure.

Figure 9. Selectivity of P ($\alpha = k_2 / k_1$).

One reactant feed is set on flow control ($f_{A,0}=1$), while the feed of the second reactant ($f_{B,0}$) is used to control its inventory. An interesting aspect is the minor variation of selectivity at higher Da (Fig. 9). Therefore, the reactor size and reactor type are ineffective for manipulating the selectivity in R-S-R. Selectivity is dependent only of the fresh feed flow rates ratio: $S_{P/A} / S_{R/A} = (2 \cdot f_{B,0} - 1)/(1 - f_{B,0})$, where $f_{B,0} \in (0.5, 1)$. This is in contrast to stand-alone reactors, where yield and selectivity reach maxima for certain values of the reactor volume. In addition, the ratio of kinetic constants can still be used to manipulate the selectivity. In R-S-R, selectivity targets can be achieved by manipulating the recycle rate, its purity, and the reactants ratio, at the reactor inlet.

Plantwide Control of R-S-R

By plantwide control we mean the strategy of solving the main dynamic problems of a process: component inventory, management of energy and safety. Plantwide control of reactant inventory can be achieved by self-regulation and regulation-by-feedback.

Self-regulation implies that component inventory is not measured, so no effort is made to regulate it by changing streams or process conditions. Implicitly, the reactants are fed in the stoichiometric ratio and the reactor is sufficiently large to transform the entire amount of reactants supplied to the system. Recycle systems using self-regulation are characterized by: 1) a feasible state exists only if the reactor volume exceeds a critical value, 2) if the reactor is small, the steady state exhibits high sensitivity to production rate changes (snowball effect), and 3) multiple steady states are possible, some of them being unstable. A major advantage is the direct set of production rate by fixing the reactant feed rate. However, non-linear effects may disturb the operability and stability.

On the contrary, in *regulation-by-feedback* the inventory of each reactant is evaluated by direct or indirect measurements, and then adjusted by changing the corresponding fresh feed flow rate. Thus, the reactor-inlet flow rates of both reactants are fixed and fresh feed rates are used to control the inventories. The strategies based on regulation-by-feedback are more difficult to implement since it requires measurements of component inventory. However, they provide two ways for changing the production rate: reactor-inlet flow rates (case of large reactors) and reaction conditions. In addition, undesired phenomena such as unfeasibility, state multiplicity and instability are avoided. This overcomes the disadvantage of setting the production rate indirectly.

The selection of the proper structure is an important step, since it determines the non-linear behaviour of the system and can provide quickly an acceptable design. These methods can be combined to find effective plantwide control structures (Bildea, 2003).

Design by Non-linear Analysis

By taking into account the design and control issues mentioned earlier, we developed a straightforward design methodology that allows the detection, at an early stage, of non-linear phenomena that may occur in recycle systems. This approach allows the designer to identify operating points that ensure stable behaviour for the desired performance, while avoiding high sensitivity faced with disturbances and parameter uncertainty.

The analysis considers a detailed reactor model while the separators can be treated as *black-boxes* that supplies constant purity specifications with variable input/output flows.

The model complexity is reduced significantly by considering separation units operated in a closed-loop fashion. The balance equations are written in dimensionless form to ensure a large range of applicability (Kiss, 2003).

Fig. 10 shows the main steps involved. The method is based on a simple plant model that captures the interaction between units and allows the prediction and exploration of major non-linear phenomena. The strategy of feeding the reactants is a prerequisite and it should be decided before performing the detailed design.

Detailed modelling is possible using process simulators. From our practice, even in complex cases the accuracy of the full model is in good agreement with the simple one (Kiss et al, 2003).

More rigorous methods including economics and operability can be used as complement (Marquardt, 2004).

Figure 10. Design methodology.

The methodology proposed in this study allows the quick screening of feasible designs and selection of promising alternatives at an early design stage.
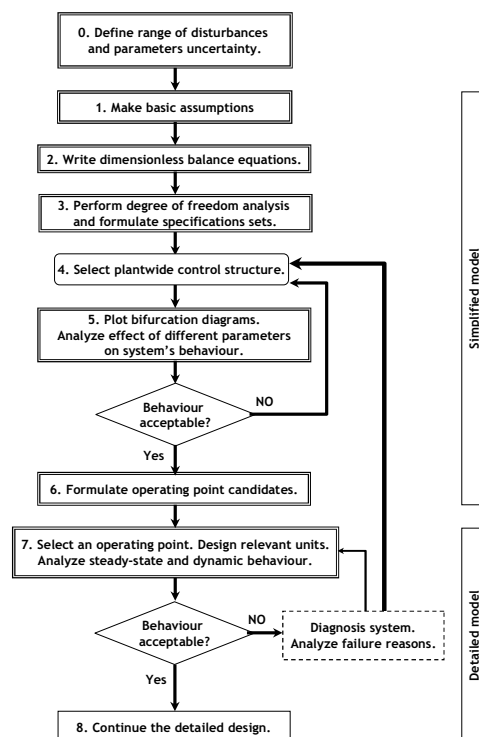
Conclusions

☑ The behaviour of recycle systems depend on the reactor size and plantwide control structure. The reactor volume must exceed a critical value for feasible operation.

☑ Snowball effect is a steady state phenomenon, so it is rather a problem of design than of control. From the control viewpoint, larger reactors behave better than smaller ones.

☑ Contrary to stand-alone reactors, in recycle systems the selectivity can be simply set from the initial ratio of reactants, independently from the reactor type or design.

☑ The results were integrated into a new methodology for design and control of recycle systems that allows the detection of non-linear phenomena at an early conceptual stage.

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