Design and control of recycle systems by nonlinear analysis
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Conclusions and suggestions for further research

The goal of this thesis was to investigate the design and control of recycle systems, by non-linear analysis. Definition of a plant Damköhler number allows the generalization of the results in dimensionless form. Unlike the classical definition of the Damköhler number that uses reactor inlet flow rate, the plant Damköhler number uses the flow rate at plant input as the reference value ($Da = k\cdot V \cdot F_0^{-1} \cdot C^{n+1}$). For recycle systems this definition is more appropriate than the classical definition of Damköhler number.

In contrast to stand-alone reactors, in Reactor – Separator – Recycle systems a zero-conversion, infinite-recycle steady state always exists if the control structure is based on self-regulation of component inventory. This state is stable if the reactor volume is below a critical value. for a given feed flow rate and reaction kinetics. Feasible steady states are possible only if the reactor volume exceeds the critical value. This constraint can be expressed conveniently using the plant Damköhler number: $Da > Da_c$. The condition corresponds generally to a bifurcation point of the mass balance equations. For example, for first-order reactions with pure product and recycle the feasibility condition is simply $Da > 1$ (Bildea and Dimian, 2000; Kiss et al., 2002). Similar expressions hold for complex stoichiometries (see Chapter 4).

Feasible states can occur by two mechanisms:

1. A transcritical bifurcation, where the infinite-recycle state loses stability and a non-trivial state gets meaningful values and gains stability. The simple one reaction systems belong to this category.

2. A fold bifurcation (turning point), at which two steady states are born. This behaviour is generic for consecutive and/or parallel reactions systems involving two reactants.

Multiple steady states are possible, even in the case of a simple reaction scheme and isothermal operation. In the bifurcation diagrams showing state multiplicity, the low-conversion states are unstable. This has practical importance only if the turning point is situated at higher conversions, for example slow termination or gel effect in case of polymerization. In this case, one cannot obtain low conversion, which might be desirable for product quality reasons, in a CSTR – Separator – Recycle system operating at a stable
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operating point. The instability of the low-conversion branch sets a lower limit on the achievable conversion.

The behaviour of recycle systems is determined by the reaction stoichiometry, recycle policy and control structure, and not by the reactor type as one would may presume. For example, the behaviour of PFR recycle systems is identical to the one previously found in similar isothermal systems involving a CSTR. The agreement refers not only to qualitative features, but also to the parameter values at which different bifurcation phenomena occur.

When heat-effects are included (Chapter 2/3, S3 and S5), one fold point may enter the feasible range of positive conversion leading to state multiplicity. If two reactants are involved in consecutive-autocatalytic reactions (Chapter 2/3, S4 and S6), state multiplicity is a generic feature. The multiplicity of states is important because it is accompanied by instability of the low-conversion branch that sets a lower limit on the achievable conversion. For isothermal Reactor – Separator – Recycle systems involving complex stoichiometry such as polymerization, this behaviour has practical importance when the radicals’ quasi steady state approximation is not valid (i.e. slow termination, gel-effect). In contrast, for non-isothermal systems the large heat effect of polymerization reactions renders multiplicity to be very probable. When designing isothermal or non-isothermal PFR-separator-recycle systems one must be aware of the non-linear behaviour and its implications on plant operability.

In practice, designs near the bifurcation points are dangerous, since changing operating conditions or uncertain design parameters can lead to a behaviour that is different from the expected one. Typically, a reactor design close to the transcritical bifurcation is unlikely due to the very small value of conversion. However, when a high-conversion fold point exists, an optimisation procedure might suggest a reactor with minimum volume. High sensitivity is likely to occur near the fold. Therefore, such a design, close to the fold, can suffer from serious operability problems. If the reaction rate is over-estimated, or the feed flow rate deviates from the nominal design value, the operating point falls at the left of the turning point in the $Da - X$ map, in the region where no steady state exists. Infinite reactant accumulation occurs in this case, and the plant has to be shut down. Therefore, in order to avoid state instability and/or high sensitivity, designs around fold must be avoided.

Self-regulation is a convenient plantwide control strategy that works well for simple reactions. However, in case of complex stoichiometry the situation is more complicated. Control structures involving self-regulation are feasible only if there are sufficient reactions to
adjust the consumption rate of each reactant such that no accumulation occurs. This condition is simple expressed by equation 4.3 (Chapter 4), which states that it is possible to put all feed flow rates on flow control only if the rank of the stoichiometric matrix is higher than or equal to the number of reactants. If the control structure implies self-regulation of one reactant, state multiplicity could occur, some states being unstable. The low-conversion branch is always unstable. This instability sets a hard constraint on the selection of the operating point. The high sensitivity of selectivity around the turning point is an additional non-linear effect in the case of parallel reactions (two recycles system). In order to enlarge the feasibility region and avoid unstable states or regions where no steady states exist, some basic guidelines are provided, as follows:

- The recycle should contain only a very small amount of product or no product at all.
- The recycle rate should be as high as possible, limited only by the economical trade-off.
- Depending on the product of interest the kinetic parameters could be changed in such a way that the selectivity is maximized. The ratio of reaction rate constants, used to manipulate the selectivity, can be adjusted by changing the reaction conditions (e.g. temperature) or using another catalyst.

Ignoring the steady state multiplicity during the design stage can lead to control difficulties. The nonlinear analysis is an appropriate tool to identify and avoid such dangerous situations at the conceptual design stage. The above mentioned guidelines are most helpful in the case of control structure consisting of setting the fresh reactant stream on flow control. If the situation permits, a stream in a recycle loop should be on flow control. This leads to stable controllable plantwide control structures, with the inlet of the chemical reactor being an appropriate location for fixing the flow rates. This location ensures stable behaviour and avoids the undesired non-linear effects.

The interaction between design and control can be revealed by mathematical models that considers the nonlinearities existing in nearly every process. The use of linear models is not excluded but linearisation around several operating points is required. The operation near bifurcation varieties, such as folds, should be avoided. If operation near bifurcation points is required due to performance reasons, the control system must ensure that possible disturbances do not lead to a catastrophic change of system's behaviour. Two basic types of reactant inventory control are possible: 1. Self-regulation of component inventory: this
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Conventional control structure consists of setting one or more fresh reactant streams on flow control. 2. Regulation by feedback control: the component inventory is measured and an appropriate feedback control system is implemented. An appropriate location for fixing the flow rates is the inlet of the chemical reactor. Feed policy of the reactants is the main difference between these two control strategies. The conventional strategy, based on the concept of self-regulation, offers more advantages but one must be aware of the non-linear behaviour when designing such systems. A major advantage of this conventional control structure is the opportunity to set directly the production rate and fix the product distribution.

The regulation by feedback control approach leads to systems where the reactor behaves as decoupled from the rest of the plant, similar to a stand-alone reactor. Therefore, conventional control strategies developed for stand-alone reactors can be applied. However, this approach is not always feasible or very helpful due to the process constrains or because the stand-alone reactor itself exhibits state multiplicity. The main inconvenient of the regulation by feedback control structure is that the production rate can not be set directly.

As simple methodology to integrate the design and control we recommend generation of design alternatives only after the nonlinear analysis is performed: identify and choose design variables; formulate controllability criteria; identify feasible and unfeasible regions. The selection of a suitable design can be made after the controllability analysis is performed for all design alternatives considered.

The original contributions presented in this thesis cover an important area of the design and plantwide control of recycle systems. Further research could consider other reactor types, reactions networks, heat-integration, different separations and product specifications. Other interesting issues worthwhile mentioning are:

- Finding an optimised method for selecting feasible control structures among various control alternatives available. The number of possible control structures is given by the different sets of specifications resulting from the degree of freedom analysis of the steady state model.

- Checking the applicability of chemical reaction network theory (pioneered by Martin Feinberg) to recycle systems. Potential extensions of the current theory would be very helpful.

- Investigating systematic methods for detecting if product distribution is fixed when all reactants are self-regulating.