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MINLP Synthesis of Reactor Networks Based on a Concept of Time Dependent Economic Regions

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Abstract

The use of economic criteria in the construction of an Attainable Region gives rise to the transformation of the conventional Concentration Attainable Region (CAR) into different Economic Regions (ERs). The more the objective function is discrete, nonlinear and nonconvex and the more its cost coefficients vary with time, the more ERs decline from the CAR. However, economically optimal solutions always lie at the borders of ERs. Some important insights have been gained during the construction of different time dependent ERs, which can be useful in upgrading the MINLP approach to the synthesis of reactor network in overall process schemes.

1. Introduction

Currently, the following three advanced approaches to the synthesis of reactor networks are:

i) The superstructure optimization approach, based on the synthesis of reactor networks, which could be efficiently integrated into an overall process scheme (e.g. Kokossis et al., 1994).
ii) The geometric approach based on the attainable region (AR) theory, which was first introduced by Horn (1964). Trajectories of a plug flow reactor (PFR), a continuous stirred tank reactor (CSTR) and their combinations are basic constitutive elements for an attainable region for 2-D problems (Hildebrandt et al., 1995) and, in addition, a cross flow reactor (CFR) for multi-D problems (Feinberg et al., 1997). Recently an automated technique for determining a candidate attainable region was proposed, for 3-D problems (Kauchali et al., 2004).
iii) A combined targeting approach which exploits the advantages of the former approaches (e. g. Lakshmanan et al., 1996).

Since AR is defined in the concentration space (CAR) using technological criteria (conversion, selectivity, ect.) rather than economic criteria, the solutions may not be economically optimal, not even in the structure. The concept of an economical attainable region (EAR) was introduced to overcome the drawback by developing a combined MINLP/EAR approach (Iršič Bedenik et al., 2004). Although it helps to find an optimal solution, it inherently involves two deficiencies:

- EAR, which is based on the transformation of CAR by economic objective function, can only be constructed consistently when the objective function is linear. However, objective functions in engineering are usually discrete, nonlinear, and nonconvex.

- Since cost coefficients in the objective function vary over time, EAR, unlike CAR, is not a static space – it is a dynamic one.

2. Construction of different ERs

In respect to the first deficiency, different types of objective functions (1a, b, c) have been studied and different reactor trajectories have been drawn to construct different economic regions (ERs) in economic space.

$$\text{Max} \quad P \begin{cases} \text{linear obj. f.:} & P = \mathrm{G}P(x) - \sum\limits_{i \in \text{reactor}} C_i^{\text{rct}} \cdot V_i & \text{(1a)} \\ \text{linear discrete obj. f.:} & P = \mathrm{G}P(x) - \sum\limits_{i \in \text{reactor}} \sum\limits_{j \in \text{size}} \left(C_{\text{fix},ij}^{\text{rct}} \cdot y_{ij} + C_{\text{var},ij}^{\text{rct}} \cdot V_{ij} \right) & \text{(1b)} \\ \text{nonlinear discrete obj. f.:} & P = \mathrm{G}P(x) - \sum\limits_{i \in \text{reactor}} \sum\limits_{j \in \text{size}} \left(C_{\text{fix},ij}^{\text{rct}} \cdot y_{ij} + C_{\text{var},ij}^{\text{rct}} \cdot V_{ij}^{C_{\text{exp},ij}^{\text{rct}}} \right) & \text{(1c)} \end{cases}$$

s.t.

$$h(x) = 0$$
$$g(x) \le 0$$
$$\left. \begin{array}{l} V_{ij}^{\text{LO}} \cdot y_{ij} \le V_{ij} \le V_{ij}^{\text{UP}} \cdot y_{ij} \,, \forall i \in \text{reactor}, j \in \text{size} \\ \sum\limits_{j \in \text{size}} y_{ij} = 1 \wedge y_{ij} \in \{0,1\}^m \,, \forall i \in \text{reactor} \end{array} \right\} \text{in cases of 2b and 2c}$$
$$x \in X \subset R^n$$

where $\mathrm{G}P(x)$ represents pre-tax profit (k$/yr), C_{fix}, C_{var} and C_{exp} are cost coefficients for investment costs for reactors, V is volume of reactor (m^3) and y is discrete variable.

2.1 Illustrative example

Let us consider autocatalytical reaction (Levenspiel, 1999) A + P $\rightarrow$ P + P, with reaction rate vector $\boldsymbol{R}(\boldsymbol{x})$, given by Eq. (2) and cost coefficients by Table 1.

$$\boldsymbol{R}(\boldsymbol{x}) = -\mathrm{k} \cdot c_{\mathrm{A}} \cdot c_{\mathrm{P}} \tag{2}$$

$c_{\mathrm{A0}} = 0.99$ mol/l, $c_{\mathrm{A}} = 0.1$ mol/l and $c_{\mathrm{A}} + c_{\mathrm{P}} = 1$, assumning that in $k = k_0 \cdot \exp(-E_{\mathrm{A}} / R \cdot T)$, $k_0 = 0.1797$ l/(mol $\cdot$ s), $E_{\mathrm{A}} = 20$ kJ/mol.

Table 1: Cost coefficients for illustrative example.

	Component A		Component P
Cost of component ($/mol)	7.34		13.90
	Cold utility		Hot utility
Utility cost ($/TJ)	981		2 614
Cost of electricity ($/kWh)	0.04		
Reactor cost coefficients ($)	$C_{\text{var}} = 8.061$	$C_{\text{fix}} = 121.530$	$C_{\text{exp}} = 0.887$

2.1.1 Construction of multi-D CAR and the corresponding EAR

The example has four degrees of freedom: τ, T, p, and the recycle rate of the recycle reactor and corresponds to multi-D problems. Any restriction in the CAR geometrical approach to 2-D and 3-D problems is overcome by one-parametric NLPs or MINLPs. One-parametric optimization with, for example residence time as variating parameter, are performed to construct trajectories of different reactors and their combinations in an attainable region. In this way 2-D projection, i.e. plots of optimization criterion vs.

variating parameter, are constructed, in which all additional dimensions are treated as optimization variables. When optimization criteria is technological, like conversion X_P in our example, trajectories define the 2-D projection of the CAR with an optimal solution of $X_P = 0.99975$ (Fig. 1). When the criteria is economic, annual profit P described by the linear objective function (1a), the procedure gives rise to the projection of an EAR (Fig. 2) with an optimal solution of P = 1949 k$/yr. However, since CAR and EAR use different criteria, their solutions are different. The construction of a multi-D attainable region is based on the procedure of one-paremetric NLP or MINLP optimization (Iršič Bedenik et al., 2004). Both approaches CAR and EAR are similar, in that, the optimal solution of the reactor network will be on the boundary of the region. In the case of CAR, the maximal X_P^{max} was obtained at the upper bound of τ, while at the optimal solution of EAR at $\tau = 13.7$ s the corresponding X_P^{opt} is 0.99925. CAR provides an answer about what is technologically attainable and EAR what is economically optimal. Based on the X_P^{max} from CAR and X_P^{opt} from EAR we can define a kind of efficiency index for a reaction system:

$$I_{eff} = X_P^{opt} \big/ X_P^{max} \tag{3}$$

Note also that, other performance criteria can be used to define I_{eff}. In our case $I_{eff} = 0.99925 / 0.99975 = 0.9995$. Since an economical solution due to cost coefficients is time dependent, I_{eff}, as will be shown later, it also varies with time. In this way information from CAR and EAR can be used to estimate current and future optimal performances of the processes.

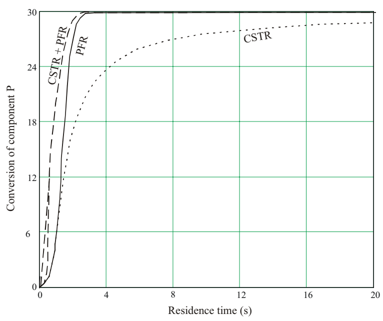

Figure 1: CAR for illustrative example.

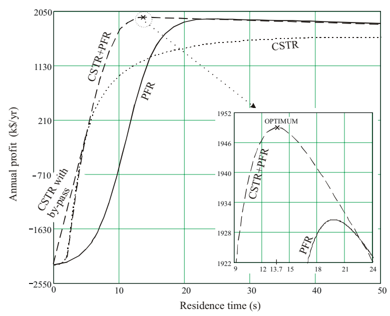

Figure 2: EAR for illustrative example.

2.1.2 Construction of different ERs

As mentioned above, if economic objective function is linear, the conventional CAR (Fig.1) is transformed into EAR (Fig. 2). However, when objective function is non-linear or continuous, the emerging economic region is no longer attainable. When, for example, the linear objective function is extended by fixed cost terms (1b), EAR is converted into an economic discrete linear region (EDLR), and when it is further extended by nonlinear terms (1c), EDLR is transformed into a discrete nonlinear region (EDNLR). The more the objective function is discrete, nonlinear and nonconvex, the more the corresponding ER declines from the CAR, and the more the principles for the construction of such spaces differ from the ones of CAR. Some similarities/differences and advantages/drawbacks of CARs and the ERs are outlined in this paper. To make the distinction between CAR and

ER clear, ER will only be called an attainable region (EAR) when the objective function is linear. The most important favorable feature, common to all ERs, is that economically optimal solutions always lie on the border of the regions. One-parametric MINLP of the illustrative example, now with a more exact linear discrete economic objective function, was performed for different reactor types to create EDLR, which also includes the trajectory of a one-unit recycle reactor (RR). Note that the fixed costs of one-unit reactors are smaller than the cost of a reactor combination that gives the same technological effect (RR vs. combination CSTR+PFR). Consequently, the optimal solution and boundary of EDLR is defined by RR. The recycle ratio must be taken as an additional degree of freedom in order to achieve an economic optimal solution. When the example was solved by the most exact nonlinear and discrete objective function (1c), the EDNLR was obtained (Fig. 3) and the optimal solution was 1875 k$/yr. It can be seen in Fig. 3, that in this case EAR and its optimal solution are slightly overestimated, while the one of EDLR is underestimated, which clearly indicates that the type of objective function effects the shape and accuracy of the ERs.

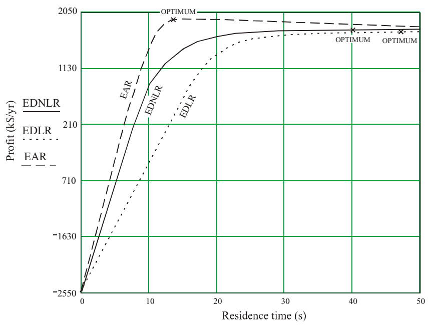

Figure 3: Boundaries of ERs.

2.1.3 Time dependency of EAR

In order to overcome the second deficiency – the time dependency of the ERs, the economic parameters in objective function terms, must be analyzed for a longer time period. A family of ERs or optimal values can be merged on a single plot where, for example, an optimal conversion or I_{eff} vs. varied investment cost coefficient can be presented (Fig. 4). The optimal conversion and I_{eff} thus dependents on the actual values of the cost coefficients. Both increase when the global economy improves and the coefficients decrease. The other possibility is for a forecasted cost coefficient (8.25 k$/yr over the first 3 years, 7.93 k$/yr over the next 4 years and 6.78 k$/yr over the next 3 years) to plot annual or efficiency index against time (Fig. 5).

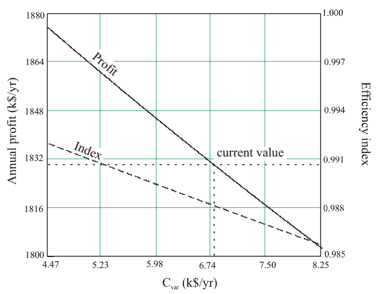

Figure 4: Conversion and efficiency index dependent on variable cost.

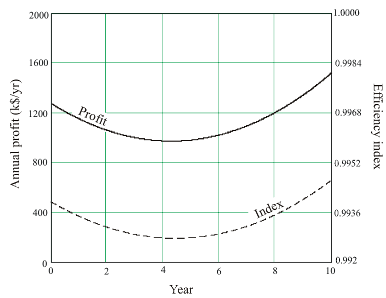

Figure 5: Annual profit and efficiency index of illustrative example.

Clearly, the values of cost coefficients affect the optimal solution. Therefore, rather than perform the synthesis of a reactor network at fixed cost coefficients, a multiperiod stochastic MINLP approach is introduced for the entire process life time to consider the varying economic parameters. For longer time periods, we use net present value (NPV) for the objective function (eq. 3).

$$\text{Max} \quad NPV = \sum_{p \in \text{period}} p_p \sum_{pp \in \text{level}} p_{pp} f_{p,pp}\left(x_{p,pp}, d\right) - \left(C^{\text{rct}}_{\text{fix},p,pp} \cdot y + C^{\text{rct}}_{\text{var},p,pp} \cdot d^{C^{\text{rct}}_{\text{exp}}}\right) \tag{3}$$

s.t.

$$h_{p,pp}(x_{p,pp}, d) = 0$$
$$g_{p,pp}(x_{p,pp}, d) \le 0$$

When the cost coefficient is fixed for the whole process life time of 10 years, the optimal NPV is 7860 k\$. In the case of the stochastic multiperiod MINLP optimization with cost coefficients defined for each period and probability level, an optimal expected NPV in the amount of 9761 k\$ is obtained. The optimal solution for the reactor networks is now time dependent and more exact than the one where the economic trends are not considered.

3. ER/MINLP Superstructure Approach for Longer Time Periods

The procedure of constructing economic regions by one-parametric MINLP is applied to the industrial problem of allyl chloride manufacturing as described (Kravanja et al., 2003) by two consecutive reactions $\mathrm{A} + \mathrm{Cl_2} \xrightarrow{k_1} \mathrm{B} + \mathrm{HCl}$ (principal one) and $\mathrm{B} + \mathrm{Cl_2} \xrightarrow{k_2} \mathrm{C} + \mathrm{HCl}$ and one parallel reaction $\mathrm{A} + \mathrm{Cl_2} \xrightarrow{k_3} \mathrm{D}$ ($k_{1,0} = 1.5 \cdot 10^{-6}$ s^{-1}, $k_{2,0} = 4.4 \cdot 10^{8}$ s^{-1}, $k_{3,0} = 100$ $l \cdot mol^{-1} \cdot s^{-1}$) with reaction rate vector $\boldsymbol{R}(\boldsymbol{x})$:

$$\boldsymbol{R}(\boldsymbol{x}) = \left[-k_1 c_{\mathrm{A}} \cdot c_{\mathrm{Cl}} - k_3 c_{\mathrm{A}} \cdot c_{\mathrm{Cl}},\; k_1 c_{\mathrm{A}} \cdot c_{\mathrm{Cl}} - k_2 c_{\mathrm{B}} \cdot c_{\mathrm{Cl}},\; k_2 c_{\mathrm{B}} \cdot c_{\mathrm{Cl}},\; k_3 c_{\mathrm{A}} \cdot c_{\mathrm{Cl}},\; -k_1 c_{\mathrm{A}} \cdot c_{\mathrm{Cl}} - k_2 c_{\mathrm{B}} \cdot c_{\mathrm{Cl}} - k_3 c_{\mathrm{A}} \cdot c_{\mathrm{Cl}}\right] \tag{4}$$

The objective is to maximize annual profit at a fixed production of allyl chloride (7.560 mol/s). One-parametric MINLP with residence time as variating parameter is performed for the whole superstructure (Fig. 6) which contains one-unit RRs and with side streams it also resembles a cross flow reactor (CFR).

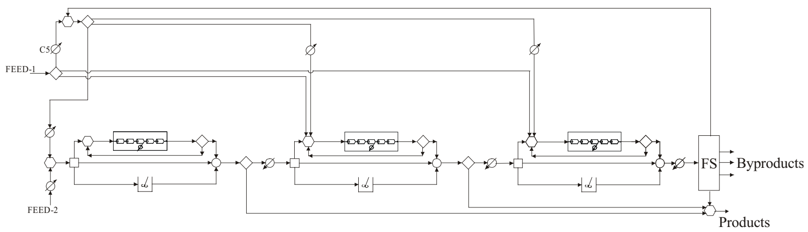

Figure 6: Reactor superstructure for industrial case.

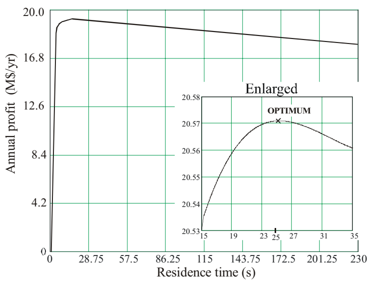

Figure 7: Boundary of economic region.

In this way the border of EDNLR is obtained directly (Fig. 7) without constructing the region by reaction trajectories. Optimal solution at 25.1 s is20.571 M\$/yr. The corresponding 2-D projection of allyl chloride selectivity with respect to chlorine is plotted in Fig. 8. If the optimization criteria was selectivity, the maximal value would be $S^{\max} = 1$ obtained at infinite recycle of the unreacted reactants. Since optimal selectivity at $\tau = 25.1$s is $S^{\text{opt}} = 0.9049$, the corresponding efficiency index of reaction system $I_{\text{eff}} = 0.9049$.

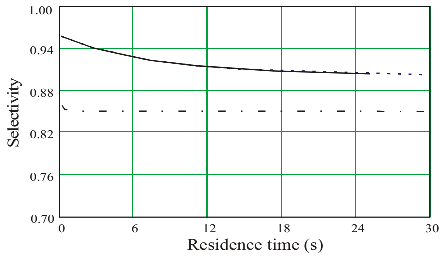

Figure 8: Selectivity on the border of ER.

3.1 Using time dependent EAR for an industrial problem

Economic parameters in objective function terms can be analyzed for a longer time period. A family of optimal values for changing cost coefficient can be merged on a single plot where P and I_{eff} vs. time can be presented (Fig. 9). With the decreasing cost coefficient term's P and I_{eff} decrease too and vice versa. Cost coefficients for past and present time periods are known and their analysis gives an exact optimal solution. However, analysis for the future is difficult because cost coefficient are uncertain. NPV for 10 years of process life period and time-unvaried cost coefficients is 83.8 M$ and I_{eff} is 0.9421. When multiperiod stochastic MINLP was performed in 3 time periods over 10 years and 3 probability levels for each period, the expected NPV of 107.5 M$ and I_{eff} = 0.9651 were obtained. The approach was implemented in MIPSYN, a successor of PROSYN, Kravanja & Grossmann, 1994. A multiperiod model contains 10230 variables, 48 discrete variables and 10089 equations, execution time was 3.93 s CPU time on a 2.5 GHz Pentium processor.

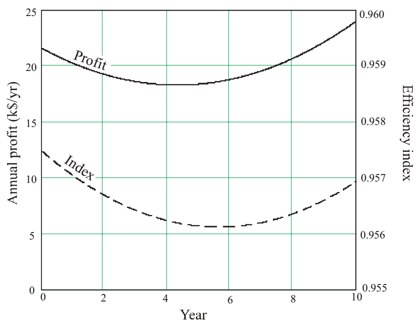

Figure 9: Changes of P and I_{eff} over process life time.

4. Conclusion

The combined targeting MINLP approach was upgraded by the principles of economic regions and by considering the time variability of the economic parameters. It was shown that if the cost trends are decreasing, the process becomes more efficient, produces less waste and becomes more sustainable. When processes are optimized for a longer period the time variability and uncertainty of economic parameters should be considered through multiperiod stochastic optimization.

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