## New Optimization-Based Approach to Process Synthesis – towards the Full Integration of Process Design, Operation and Control

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Conceptual process design still follows a traditional procedure which consists of three sequential stages, as depicted in Figure 1. In the first stage, the operation mode is selected according to the particular process and manufacturing demands. The decision is strictly based on heuristic rules, for instance batch operation is preferable for small capacities and multi-products while continuous operation is recommended for bulk production of a single product. In the second stage, the process is designed according to the process' physical and chemical characteristics, desirable production rate and quality and related economical issues. Although in this step equipment dimensions are commonly calculated using mathematical models, this phase also heavily relies on heuristics. In most cases predefined process units for a specific application are used and a selection of units is completed according to this practical method. In the third phase, a control system is designed for the purpose of maintaining the specified product quality and quantity, as well as process safety. This procedure is well established in practice; it offers ready-made mathematical models and technical solutions. However, it does not allow the interaction of design, operation and control and therefore it is generally suboptimal from both economical and environmental perspectives.

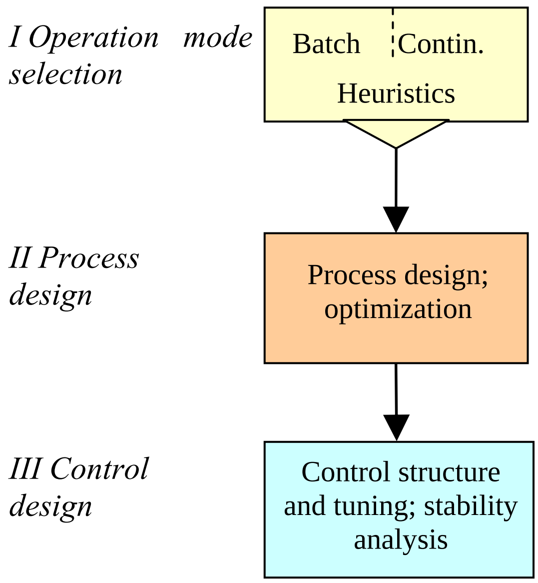


*Figure 1. Traditional process synthesis*

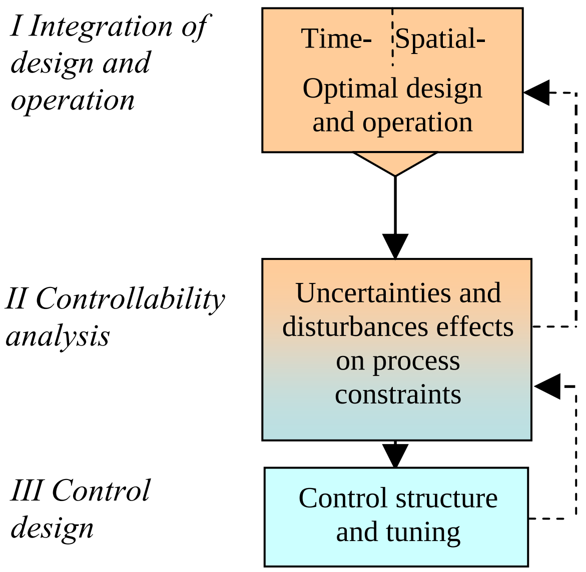


*Figure 2. New optimization-based process synthesis*

A new approach to chemical process synthesis is going to be presented and illustrated on an example. The concept is based on dynamic optimization and it also contains three stages as presented in Figure 2. In contrast to traditional approach this concept provides strong interactions between process operation, design and control. In the first stage, the complete integration of process design and operation should be attained by means of dynamic optimization, based on simple first principles mathematical models. Optimization objectives are based on economical modeling and they may include environmental issues. Optimization results should provide optimal process design and optimal operation. Proposed process synthesis concept is it is out of the limits of traditional unit operation approach and it is determined by exploration of phenomena and manipulation of fluxes and driving forces. It should be emphasized that the operation mode is not fixed *a priori* (as in Fig. 1) but is a result of the optimization. Therefore, this concept promotes the exploitation of process intensification principles and methods for process design. It also explores the possibilities for actuation improvement for optimal operation and control.

In the second stage (see Figure 2) controllability study for the optimal process solution (achieved in the first phase) is to be performed, by means of robust optimization techniques. In this analysis, the effects of model uncertainties, specifically physical parameters uncertainties, and process disturbances on the process constraints are to be examined. This analysis may lead to adjustment of actuation variables, in order to avoid violation of the constraints. In the third phase, the control system is conceptually designed, taking the controllability results into account. Its target is to follow the optimal operation path as effectively and robustly as possible. It is reasonable to assume that existing advanced control systems, like (nonlinear) model predictive control, or optimal control, will function adequately for the optimal designs and realization of the optimal operation. A closed loop information is transferred back to the stage two (dashed line) in order to perform closed-loop controllability analysis, which could eventually lead to additional actuation alteration (dashed line to the first stage).  In this presentation, the first and most important stage of new process synthesis concept will be addressed, while the second stage will be tackled in the following paper.

The methodology for attaining the integration of process operation and design (stage one in Figure 2), could be generalized as follows. First, after a chemical problem is given (with known reaction mechanism and properties) an economic objective function is modeled, which includes the revenues for a production of desired product and costs for: a) raw materials b) investments, c) energy, d) environmental issues. The problem is then systematically analyzed to consider as much as (physically) possible structural and operational enhancements, in order to maximize fluxes and driving forces within the process. For that reasons various process intensification methods are considered. Formulation of a single unique mathematical model which incorporates different process structure options and ways of operation is not attainable at the present. Therefore, the proposed method implies the decomposition of a generic problem into two or more sub-problems to be subjected to an optimization (Figure 2). One type of general model will generate more spatially-oriented solutions, operated often in continuous regime, both steady and unsteady state. The second form of model will result in more timely-oriented solutions, operated more frequently in discontinues or semi-continuous mode. After the concurring problems are selected and models are set, all degrees of freedom (control variables) need to be listed and classified between time-variable or time-invariant. Apparent physical constrains are to be incorporated in the optimization set-ups, as usual. After dynamic optimizations (MIDO) are performed, the target function values will be compared for a selection of the more beneficial solution, which would further be treated in stages two and three.

Proposed process synthesis methodology, precisely its first phase, is illustrated on an industrially relevant example – methane steam reforming (MSR) accompanied with water gas shift reaction (WGS). The goals are to have: a) high conversion of methane; b) efficient separation of hydrogen; c) high driving forces for reaction and separation; d) low energy costs; e) low capital costs and f) compact design. To overcome the equilibrium limitations and achieve high conversion of methane, a functional integration of catalytic reactor with $H_2$ selective membrane is a very good process intensification option. A membrane will also provide immediate separation of a product from the undesired product and allow the use of lower temperatures. The alternative could be an integration of catalytic reaction with *in situ* adsorption; however this option was not consider at present, in order to have manageable and illustrative example. To attain high driving forces for the membrane separation, as well as for the reactions, consecutive order of process functions in one unit (reaction-separation-reaction-…) is considered, aside to fully integrated system. Without going into technical details, the spatially-distributed continuous systems could be realized as a tubular catalyst bed reactor containing a number of membrane-wall tubes for the $H_2$ permeate, if the process functions are integrated. The other, time-distributed class of systems could be designed as a rotating bed reactor with membrane-wall tubes. To minimize diffusion resistances, a structural packing with Ni catalyst coating will be assumed. It should have a large surface area, but high bed void fraction, similarly like foam type of packing.

Alongside mentioned structural and functional intensification, the actuation improvements should be examined systematically and rigorously within the optimization. The first reaction (MSR) is highly endothermic and requires high temperatures and the second reaction (WGS) is slightly exothermic and requires moderate temperatures. In order to attain optimal reaction temperature profile a spatially distributed heat supply will be considered and optimized. The optimal heat supply, with high energy efficiency could possibly be realized by using electric current as a direct heat sources in the catalyst. Alternatively, microwaves could be used for efficient and selective heat supply to catalyst. Further, optimal control in space and time with these alternative energy sources could be achieved effectively due to fast dynamic characteristics electromagnetic fields. For the continuous system, unsteady state (periodic) operation is also examined, as another possibility for operational improvement.

Economical target function comprises a maximal production of hydrogen with a minimal costs for catalytic and membrane surfaces and energy consumption. The dynamic models are simplified using common assumptions in order to attain convergence during the optimizations with a number of constraints and control variables. Altogether, six optimization problems have been derived and analyzed. For all analyzed systems the dynamic optimization results provide optimal design parameters and optimal operation, satisfying process constraints. Optimal design parameters include: number of parallel systems, number of membrane tubes per unit, diameters of units and membrane tubes, lengths, diameter of foam pore, etc. Optimal operation parameters are: inlet steam to methane ratio, inlet temperature and pressure, inlet flowrate of sweep gas, spatially distributed heat supply, etc. The base case system for the comparison of economic performance consists of one reaction unit followed by one separate membrane unit, similarly to a conventional system. Finally, the comparison of the objective functions (and the terms related to the profit and costs) between six analyzed systems suggests the overall optimal process solution. Beside that, the comparison of reaction rates and permeation rates profiles between several optimal process solutions, provide more insightful information and grounds for the selection of the optimal solution. In conclusion, the results demonstrate significant difference in economic and technological performance. This example demonstrates the new methodology well, and provides the basis for the further development of the suggested concept.