



Opportunities and challenges for process control in process intensification

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ABSTRACT

This is a review and position article discussing the role and prospective for process control in process intensification. Firstly, the article outlines the classical role of control in process systems, presenting an overview of control systems' development, from basic PID control to the advanced model based hierarchical structures. Further on, the paper reviews the research articles discussing control issues of intensified process equipment, specifically of reactive distillation, divided wall distillation, simulated moving bed reactors and micro-scale systems. In the next section, the focus is on more fundamental, dynamic characteristics of selected intensified process categories, which are elucidated in several examples. The goal of this analysis is to stress to the potential challenges for control of intensified processes. More importantly, the aim of this part is to emphasize to the opportunities for control, which are associated with new actuation possibilities arising from process intensification. Finally, a new concept of process synthesis is elaborated, which is based on process intensification and actuation improvement. The concept enables integration of process operation, design and control through dynamic optimization. This simultaneous synthesis approach should provide optimal operation and more efficient control of complex intensified systems. It may also suggest innovative process solutions which are more economically and environmentally efficient and agile.

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1. Introduction

The process industry is confronted with diverse challenges. First of all, society requires a sustainable industrial development outlined by environmental responsibility, renewable energy use

and higher energy efficiency. These requirements are expressed in more stringent legislation regarding waste production, CO₂ emissions, air and water pollution, etc. On the other hand, economic pressures are prevailing—global competition, dynamic change of market demands, short time to market, more efficient supply chain, considerable cost reduction, etc. These economic demands and unstable economic situation are forcing industry to be more flexible and to be able to change production capacities fast. Furthermore, frequent variations in feeds' compositions, together with

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fast and sometimes unpredictable prices' oscillations give much more emphasis on process dynamics. Combined together, all listed expectations are raising many scientific and technical issues for modern process technologies and their control.

Looking at the chemical engineering success in coping with the challenges in the process industry, one could locate process intensification (PI) concepts as the most promising ones [1], especially when linked with green chemistry (GC), product design and process systems engineering (PSE). Process intensification brings the 'paradigm shift' to process design, offering novel processing methods and equipment "which can benefit (often with more than a factor two) process and chain efficiency, capital and operating expenses, quality, wastes, process safety and more [2]." Green chemistry contributes by offering new products and new synthesis routes, which are more efficient and environmentally friendly; therefore GC enables process intensification to fulfill its potentials. Process system engineering offers the tools for a systematic approach to complex problems in process design and operation [3]. PSE's major achievements include methodologies and tools to support process modeling, simulation and optimization [4], which are of major importance for process intensification. And vice versa, process intensification provides 'building blocks' for further development of PSE [3,5].

Research interest in process intensification is growing, according to the large increase in number of publications throughout the last decade [6–9]. To date, several monographic books have been published presenting the principles, modeling and applications of process intensification [10–15]. It could be concluded that PI research is approaching a mature stage. Several applications of process intensification principles are realized so far on an industrial scale. Most successful commercial PI applications include reactive distillation, microreactors, rotating packed bed systems, simulating moving bed reactors, etc. [10,12,16–18]. However, there is still a disproportion in the radical improvement potential of PI and the state of the implementation in the industry.

From a process control side, there are some research contributions discussing control aspects of intensified processes. Most of them are dedicated to reactive distillation (RD), which is understandable, since there are already over 150 industrial applications of RD [17,18]. However, almost no information on control of commercially realized intensified process systems is available in the open literature. Generally, control of PI systems is facing challenges, which are associated with the nature of intensified processes' dynamics. At the same instance PI is offering opportunities for a fundamental improvement of process operation and control.

Above mentioned demands of modern society call for further advances in process industry, next to emerging intensification of equipment and product design. Control design is a part of process synthesis (PS), typically its final phase in the traditional consecutive approach. Although this classical method is well-established in the practice, processes can be improved if control is considered simultaneously with operation and design. Furthermore, process synthesis can offer innovative and more efficient process solutions if it systematically includes various process intensification concepts.

What is the role and prospective for process control in PI? How can operation of intensified processes be moved closer to economical and environmental optimal performance with tighter constraints? Could process operation and actuation be intensified as well? In this text we are trying to answer these questions. First, we are going to present the development of process control systems, in order to outline the classical role of process control. In the following section, research articles regarding control of PI equipment are reviewed. After that, we examine dynamic characteristics of certain PI classes and summarize the consequences for operation and control. Finally, we present a new concept of process synthesis

which aims to the full integration of process operation, design and control by means of dynamic optimization. This concept exploits different process intensification methods and explores a potential for actuation enhancement. We hope that this paper will open the discussion about the new role of process control, its more logical position in chemical engineering and process industry as an active 'intelligent' component, not a passive regulatory system depending strongly on a process design, as it has usually been so far.

2. Development of process control systems and state-of-the-art

Mainly due to an almost unrestricted consumer demand the process industry expanded tremendously in the "early years" from 1950s to 1970s. During these decades the theoretical developments in process control were directed towards optimal control. Pontryagin's maximum principle, for example, was used to maximize the conversion in a tubular reactor [19]. Another important contribution came from Buckley [20] who was the first to formulate the objectives of a process from a plantwide perspective that resulted in a structural decomposition, material balance and quality control. The industry was using PID (proportional integral derivative) control including extensions like ratio, override and split-range control which was all implemented pneumatically. Although the process industry already experimented with direct digital control (DDC) in the late 1950 it was concluded that modern control techniques were complex, expensive and did not generate enough economic benefits. By the end of this period there was consensus that more profitable process operation required a hierarchical structure; a lower level regulatory control layer and an upper level steady state optimization layer.

In the mid 1970s the first oil crises caused a tripling of the fuel costs almost overnight. This gave an emphasis on energy conservation, especially in distillation. At that time approaches to reduce the energy consumption in distillation, such as heat integration at the reboiler or condenser level, vapor recompression or double composition control, were deemed too complicated from a control point of view (Luyben [21]).

The ongoing computer hardware developments paved the way for cheaper and more powerful microcomputers. As a result Honeywell and Yokogawa both introduced in 1975 their first generation distributed control system (DCS); TDC 2000 and CENTUM, respectively. Just as DDC, these systems have shown much better computational capabilities and a much higher accuracy, compared to pneumatic control. However a DCS also offers two additional advantages: (i) standardization, these systems can be configured by means of control function blocks like PID, ratio, override and split-range control (ii) reliability, a DCS consists of a number of microcomputers and each microcomputer controls 8–16 individual loops. Therefore failure of one microcomputer just means loss of a limited number of control loops and not a complete plant shut-down.

In the 1980s, due to global competition, there was an increasing appreciation for the relation between product quality and profitability. The process industry was already aware of the fact that off-specification production should be avoided. However the relation between quality and profitability goes beyond just avoiding off-spec product, see Fig. 1. Improved (e.g. advanced) process control can reduce the variability in quality or, as pointed out by Downs and Doss [22], control typically relocates variability from process output(s) to process input(s). After the variability reduction the set point of the control is increased to exploit the improvement economically. A higher set point can be associated with lower cost if the quality is expressed as an impurity level. The higher set point might even allow for a higher throughput, since higher impurity

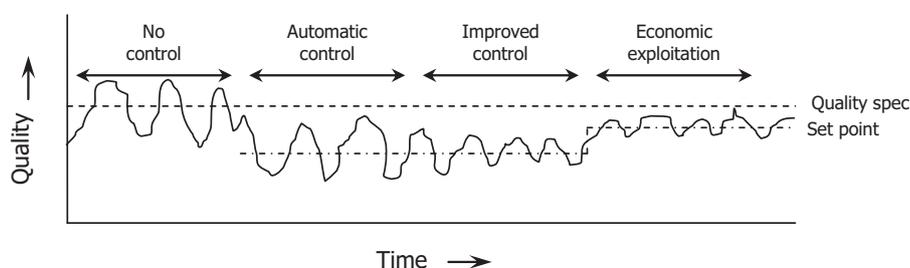


Fig. 1. The variability of quality over time during various stages of control.

Table 1
Control and optimization in state of the art continuous manufacturing.

Layer	Execution frequency	Scope	Model
6. Planning	1/week (s)	Site	Steady state linear
5. Scheduling	1/day	Site	Steady state linear
4. Real time optimization	1/h (s)	Plant	Steady state non-linear
3. Model predictive control	1/min	Unit operation	Dynamic linear
2. Base layer control	1/s	Proc. var.	–
1. Actuation and sensing	1/s	–	–

Table 2
Control and optimization in state of the art batch manufacturing.

Layer	Spatial scope	Temporal scope
4. Batch production management	Plant – site	“All” batches
3. Run to run control ^a	Unit operation	Multiple batches
2. Within the batch control	Unit operation	One batch
1. Actuation and sensing	–	–

^a Also called “batch to batch control”.

levels often translate in lower utility demands, because of a less demanding separation. In this period also health, safety and environment (HSE) issues became more important, requiring plants to be operated at maximum efficiency, satisfying clear constraints on the induced environmental load. All this led to more complex plant designs that were characterized by less intermediate storage, more and larger recycles and more extensive downstream processing. Obviously such plants are more difficult to control.

The beginning of the 1980s was also marked by the rise of model-based control, more specifically model predictive control (MPC, Cutler and Ramaker [23], Richalet et al. [24]). The success of MPC since its introduction is not difficult to explain, this control technique can deal in a complete unified way with constraints (state and input), interaction, measured disturbances (feedforward), inferential control and non-minimum phase behavior. It should be mentioned that MPC could be implemented because of the availability of sufficient computational capability at reasonable cost.

The period from 1990s to present had a particular focus on model-based decision making. This implied that process modeling, control and optimization became equally important. Since the beginning of 1990s the number of real time optimization (RTO) applications started to grow, see Marlin and Hrymak [25] for an overview. RTO basically provides the missing link between scheduling and MPC (see Table 1).

The current state of the art of the continuous processes has six layers; layer 1–3 deal with control while layer 4–6 are involved with optimization. Layer 1 provides the process with inputs and outputs. Base layer control¹ (BLC) consists of PID control including extensions like ratio, override and split-range control. The scope comprises one or two process variables. The third and last control layer has a larger scope; typically a unit operation like a reactor or a distillation column, while the model is of dynamic linear nature. The first optimization layer is RTO that basically computes the optimal set points for the BLC and MPC of a particular plant most commonly using a steady state non-linear model. The separation between RTO and MPC has become more blurred over time. Considerable research effort was directed to non-linear MPC (NMPC) enabling to cover a larger operating range, however also the objective has become more economic [26–28]. Huesman et al. [29] point

out that the limitations of RTO are related to the use of a steady-state model. As a result RTO only support continuous operation, tracks the economic optimum slowly and cannot optimize transitions from one steady state to another. The last two optimization levels are normally mentioned together (scheduling and planning; S&P) probably because they share the model. This model has a site wide scope. A site is a collection of plants that share the organization (management, technical staff, maintenance, etc.) but also things like storage and utilities. However planning typically generates economic forecasts and production goals while scheduling basically determines how to execute the chosen plan, the key issue being feasibility with respect to confirmed orders.

Batch processes have received considerably less attention than continuous ones regarding process design and control. This is clearly reflected in standard chemical engineering textbooks as well as in literature on process control. The various control and optimization layers have been described by Seborg et al. [30], see Table 2.

Layer 1 performs the same function as in continuous manufacturing. “Within the batch control” entails discrete and continuous control, which is capable of automatic recipe execution. The discrete part is often described and implemented by sequential function charts (SFCs) while the continuous part can be as simple a PID control. “Run to run control” adjusts the recipe for the next batch, in which improvement or even optimization of the recipe is done based on off-line quality measurements like chromatographic methods. The fourth and last layer schedules the various unit operations taking into account customer demand, the availability of raw material, etc. Batch production management maximizes the asset utilization; a non-trivial issue in batch production. It is quite easy to relate Tables 3 to 2. “Within the batch control”, BLC and MPC all have control functionality; the set points or set trajectories are coming from the layer above. “Run to run control” and RTO have in common that they can only perform their task at certain moments in time; after the off-line quality measurements have become available or after the plant has reached steady state. “Batch production management” and S&P in particular scheduling obviously have the same functionality.

Despite the similarities just mentioned, Bonvin et al. [31] point out a number of differences when comparing batch with continuous operation, the most important ones being the challenging task of deriving reliable (kinetic) models, the difficulty of on-line measurements, the lack of (semi) steady-state behavior, and

¹ Also referred to as regulatory control.

consequently the necessarily larger operating range which does not allow linearization of the dynamical behavior.

The development towards the widespread use of model-based control and optimization has shifted the emphasis from control aspects to modeling aspects. According to several studies (see Darby et al. [32]) it is known that in modern process control and optimization projects, the majority of effort (and budget) is spent on developing the appropriate dynamic models. Process models can be obtained either by rigorous first principles modeling or by process identification (Ljung [33], Zhu [34]). Mathematical modeling requires expertise and is labor intensive. Rigorous and detailed process models generally are rather complex and their use is often limited for following reasons:

- (a) Validation of rigorous dynamic models is not an easy task.
- (b) Their computational complexity usually makes them less suitable for on-line applications, e.g. model predictive control.

Model reduction techniques are generally applied to circumvent the second problem, but in the most common situations MPC controllers are based on (linear) process models that are identified from experimental data, as e.g. step tests or experiments with more general input excitations. While these identification methods are well developed for linear models, this linear approach limits the operating range for which the models are suited. Identification of nonlinear process models from experimental data is a topic of current research, and very much focuses on the choice of appropriate (black-box) model structures. The further development of nonlinear MPC (NMPC) very much depends on the availability of reliable and experimentally validated nonlinear process models.

With the development of efficient nonlinear MPC, the distinction between RTO and MPC slightly changes and opportunities occur for combining the economic objectives from RTO with the set-point tracking objectives of MPC [26–29]. However, the resulting dynamic optimization requires the availability of efficient (large scale) optimization methods.

Additional developments in the process control field include:

- (a) Methods for decentralized and distributed control, for processes with larger numbers of (local) actuators that are bound to jointly achieve a particular control and optimization task, under different levels of internal communication.
- (b) Design of experiments that are least costly/least intrusive.
- (c) The development of approximate linear models that are best suited for a particular model-based control objective.
- (d) Robust control design methods that incorporate a particular level of insensitivity to model uncertainties.
- (e) Multi-parametric approach for MPC which exploits the analytical solutions in particular regions of the variables (state) space in order to decrease the required on-line computational effort.

Focus of process control has been changing over the recent time since certain classes of process systems have drawn more attention, specifically:

- (a) *Fine and pharma*. The growing importance of fine chemicals and pharmaceuticals in developed areas like Western Europe has raised interest in batch control. Of course (fed) batch operation is also important for sectors like biotechnology and the food industry. Currently, a number of companies are developing processes and equipment for a continuous production of fine chemicals/pharmaceuticals instead of the traditional batch processing. Usually complex chemical and physical behavior of fine chemicals' reactions is bringing even more challenges for advanced model based control and on-line analytical tools in continuous systems.

- (b) *Energy*. The number processes under investigation related to energy is rapidly increasing. A lot of research implies switching over from oil, gas coal, etc. to renewable feed stocks like trees, agricultural crops and municipal solid waste. Renewable feed stocks are often season bound and vary in quality over time. Few more examples where energy systems are highly integrated and become more dynamic are: large scale carbon dioxide capture (post- or pre-combustion), (co)-combustion or gasification of biomass and the production of energy carriers (hydrogen, methanol and ethanol economy).
- (c) *Unmanned*. There are a growing number of process systems that must function without the supervision of an operator. Typical examples are oil and/or gas platforms, fuel cells (automotive and domestic) and integrated micro power systems.

The various classes of specific process systems indicate the growing importance of non-stationary and agile process operation, the word agile meaning fast adjustment towards new conditions (for instance other product, energy demand or feedstock). Such forms of operation must be "supported" by process and control design. In this context process intensification is particularly interesting since high fluxes lead to compact equipment with fast dynamics.

3. Control of intensified process systems—current status and role

Control of intensified process systems have been studied since 1990s with dynamics and controllability as predominant subjects. The majority of academic contributions is devoted to reactive distillation, which could be expected, as those systems have been developed and implemented for a commercial use. In reactive distillation, the interaction of reaction and separation causes complex behavior such as process gain nonlinearity, significant variables' interactions, process gain sign change and steady-state multiplicity. Moreover, reactive distillation, like other multifunctional systems, exhibit a reduction of operational degrees of freedom as well as a reduction of operational windows, which is addressed in detail in part 4. There is a considerable amount of research papers on dynamic simulation, controllability analysis, control design and interaction of design and control of reactive distillation process published in the literatures [35–45]. In these publications different approaches for control have been investigated, from traditional PI to more advanced model predictive control, both linear and nonlinear types and they will be discussed shortly in the following text. For more detailed outlook readers are referred to the recent review on control of reactive distillation columns by Sharma and Singh [35].

Majority of the research work which uses classical PI control was focused on the proper selection of the control variable(s) and the actuation variable(s) for the common reactive distillation cases, e.g. ETBE, MTBE. In several publications Al-Arfaj and Luyben [36,37] analyzed PI control structures with one- or two-point temperature control. They started with an ideal, generic two-product reactive distillation column and proposed a variety of control structures for two-point control of product purity in which the concentrations were controlled with direct measurement of compositions in the system by PI controller [36]. In another study, Al-Arfaj and Luyben [37] applied control structures developed in the previous work to an optimized double-feed ETBE reactive distillation column. They proposed a single point control PI structure controlling either the bottom product composition or a stripping section stage temperature for a single-feed ETBE reactive distillation column. Sneesby et al. [38] proposed an inferential control scheme for two-point control of ETBE reactive distillation column in which both

bottom product purity and reactant conversion are controlled using conventional PI controllers. Bartlett and Wahnschafft [39] studied the control of a methyl *tert*-butyl ether (MTBE) reactive distillation column. Several schemes using conventional PI controllers are discussed. The selection of an appropriate tray temperature is explored. The authors recognized the importance of maintaining tight control over the feed stoichiometry to avoid an excess of methanol and therefore they recommended a feedforward scheme.

Some authors argued [40,41] that a linear proportional–integral–derivative algorithm with fixed parameters is not satisfactory for handling the high directionality of the process gain in reactive distillation. Therefore for a retuning of process gain, several adaptive approaches have been proposed. Tian et al. [40] proposed a pattern-based predictive control (PPC) scheme for single-point control of the bottom product ETBE purity. They introduced a nonlinear transformation to obtain a pseudo-linear input–output relationship and incorporated the PPC with conventional PI control. Bisowarno et al. [41] proposed two adaptive PI control strategies, model gain-scheduling and nonlinear PI, to improve the single-point bottom product ETBE purity control performance. In gain-scheduling approach [41], a set of simple local models were attained, which cover relevant operating conditions and cope with the nonlinear characteristics of the process. The simple models are then integrated by using a proper switching scheme. Simulation results have shown that the proposed control strategy outperforms the standard proportional–integral control.

The other researchers suggested model predictive control for RD columns, especially for the two-point control option, because of strong interactions between process and manipulated variables. For the control of a continuously operated reactive distillation column, Khaledi and Young [42] used a 2×2 unconstrained linear model predictive control for two-point control of the ETBE reactive distillation column. The authors demonstrated [42] that MPC is able to handle the process interactions better than PI controllers.

Kumar and Daoutidis [43] proposed a nonlinear input/output linearizing controller, for which they show it is more effective than PI at moderate product purities but was unstable at higher purities for production of ethylene glycol. Similarly, Vora and Daoutidis [44] used a nonlinear input/output linearizing state feedback controller, which is tested by making setpoint changes in the distillate composition for the ethyl acetate reactive distillation process. Grüner et al. [45] derived a nonlinear state feedback controller by combining input/output linearization with the design of a robust observer. Although originally developed for their column, the control-law as well as the observer design is presented in the paper [45] in a generic formulation allowing a straightforward adoption for different reactive distillation columns.

In several contributions, a problem of high computational load of non-linear MPC was tackled with different model reduction approaches. Doyle and Balasubramhanya [46] employed the concept of travelling waves to generate reduced-order nonlinear process models which are used in a nonlinear MPC-scheme in a batch reactive distillation column producing ethyl acetate. Volker et al. [47] developed a control design procedure of a semi-batch reactive distillation column for methyl acetate production. The design procedure consists of three steps. First, a suitable control structure, that enables the operation near the economically optimal operating point, is determined using rigorous nonlinear process model. In a second step, a linear model of the column is identified from experiments and then used to compute the best attainable control performance for the chosen control structure. In the third step, the resulting high-order controller is approximated by a low-order controller that gives nearly the same performance and preserves robust stability for the examined uncertainty bounds. The controller performance is tested in a series of experiments that were performed at the pilot-scale reactive distillation column [47].

In divided wall columns, or so called thermally coupled distillation columns, energy is utilized more efficiently than in conventional distillation. Similar to reactive distillation, divided wall columns exhibit complex non-linear dynamic behavior with narrow operating windows [48–51]. Again, different control approaches were used, starting from PI control and its modifications such as PI with dynamic estimation of uncertainties (quadratic action) [49]. As expected, more authors propose optimization based control of divided wall columns as it outperforms classical PI [50,51].

Simulating moving bed (SMB) principle is commonly used in separation, as it provides continuous operation of normally discontinuous chromatography. SMB can be combined with reaction, for gaining synergetic effect of integration, e.g. overcoming equilibrium reaction limitations or achieving thermal coupling [52]. These periodically operated (columns inlet switching) and spatially distributed systems, were in the focus of several control research groups [52–54]. For glucose isomerization process, Taumi and Engell [53] used non-linear model predictive controller which can deal with complex hybrid (continuous-discrete) process dynamics. This control system was tested and proven to be efficient on the small-scale experimental system. For control of heat integrated SMB reactor, Zahn et al. [54] proposed a closed loop discrete controller based on Petri net. Three control schemes, which differ in measurement strategy (sensor numbers and locations), were analyzed and the most promising control structure was examined experimentally [54].

Control of micro-chemical systems, like microreactors or microseparators, was addressed in several published articles so far. Hasebe in his paper [55] presents design and operation issues of micro-chemical plants, where he briefly addresses control and instrumentation. Hasebe points out that the micro-plant and control systems should be designed simultaneously. In his first argument he claims that it is almost impossible to install measurement devices into micro-system, once the unit is constructed. In the second example author shows how a proper design of microreactor walls (higher conductivity) could offer more robust temperature control, less sensitive to flow disturbances [55]. Hasebe also emphasize the importance of reducing the number of control variables (outputs of many channels) by adopting an indirect control system, for instance control of heating medium temperature for the control of process temperature. In the recent publication from his group [56], Hasebe derives model-based monitoring system with Kalman filter as an observer, for fault diagnosis in micro chemical processes. Furthermore, the authors [56] discuss operational policies and control structures for parallelized micro chemical devices which are subjected to blockage of some channels. They recommend a pressure drop control over a total flow control for such operational problems and show some experimental validation [56].

Kothare and co-workers [57,58] offered advanced model-based control for micro-systems, with their particular interest for a control of microreactors for fuel processing (methanol reforming, water gas shift). The block diagram of their receding horizon feedback controller is presented in Fig. 2. The basic model was constructed using first principles equations for fluid flow (Navier–Stokes plus slip at the walls) and conservative laws for a detail geometry of the microreactor [57]. A distributed (PDE) dynamic model was solved by COMSOL Multiphysics (previously FEMLAB). For the application in on-line control, a rigorous model has been reduced to the low-order model by implementing proper orthogonal decomposition (POD). Proper orthogonal decomposition is applied on an ensemble of simulation data to obtain the dominant dynamic structures, called empirical eigenfunctions. Bleris and Kothare [58] proposed a new receding horizon boundary control scheme using the empirical eigenfunctions in a constrained optimization procedure to track desired spatial–temporal profiles

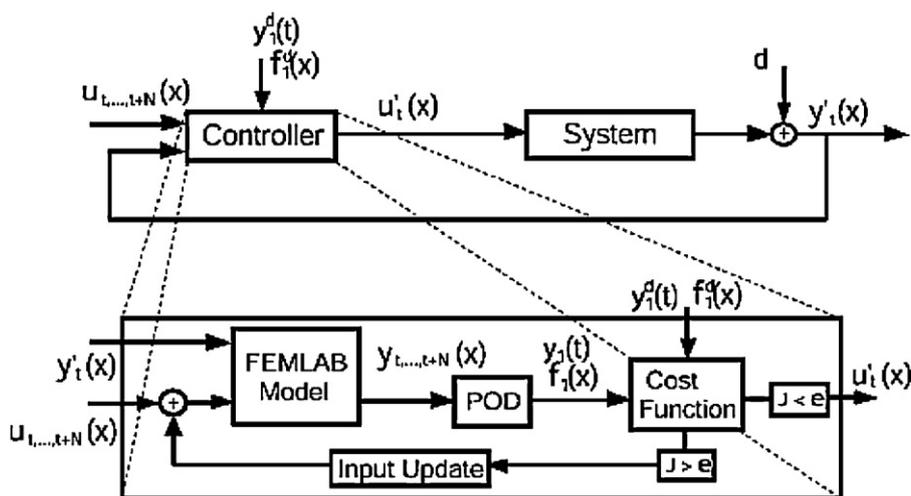


Fig. 2. Block diagram of receding horizon feedback control of microreactors.

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(Fig. 2). As shown by Bleris and Kothare [58], this controller was able to maintain a desired temperature profile in a microreactor using resistive heaters as actuators. They have also reported its successful use in regulation of flow in microchannels and in switching flow for distributing micro-flow patterns [57].

Presented overview confirms that a majority of current research interests in control of intensified process systems are oriented towards advanced model- and optimization-based control. Conventional PID systems (with modifications) are also examined as they are simple and still widely used in the industry. Further, in some cases, effort for modeling and parameter tuning could be up to three times higher for MPC in comparison to classical PID. The main technical challenges for advanced model-based control of intensified process systems are analogues to the ones for general process systems, discussed in Section 2. To reiterate, they are: time consuming modeling process, implementation of a proper model reduction scheme and realization of non-linear MPC. However, intensified processes have specific dynamic and operating characteristics. Those may raise challenges for control, but also offer new opportunities, and therefore will be emphasized and discussed in the following section.

4. Dynamic and operating characteristics of selected intensified processes categories—challenges and opportunities for process control

Throughout the following sections, process intensification approaches are assembled in four fundamental domains [5], and thus related to process operation, design and control. The PI fundamental domains and related main focuses are:

1. *Functional*: integration of process functions (multifunctionality).
2. *Thermodynamic*: alternative driving forces and energy sources.
3. *Spatial*: well defined geometry (structure).
4. *Temporal*: dynamic operation and time-related realizations.

Since this article focuses on control and operation, time domain is essential. For this reason dynamic aspects are analyzed throughout all other PI domains and not separately.

4.1. Hybrid units—operational constraints

One of the basic principles of PI is utilization of the synergistic effect, which can be effectively exploited in multifunctional

systems, combining more process operations or steps in one unit (*functional domain* of PI [5]). Alongside reactive distillation which already has industrial commercial relevance [17,18], the other combinations of chemical reaction and separation, or two separation processes in one unit are promising. However, those hybrid systems have less degrees of freedom (DOF) than classical systems with separated consecutive units. It should be noted that generally several definitions of degrees of freedom can be distinguished; among them the ones important for process systems are:

- (a) thermodynamic DOFs;
- (b) design DOFs; and
- (c) operational DOFs.

Thermodynamic DOFs define the number of intensive system properties (like pressure or temperature) which are independent of the other intensive properties. Design DOFs identify the number of independent geometrical properties, which can be used for the system design, like diameter or height of a column. Operational DOFs define independent process variables which can be manipulated for the purpose of control or desired operation. Though defined differently, DOFs are often related, for instance less number of thermodynamic DOFs reduces the options for design and control, as illustrated in the following analysis for hybrid systems.

Integration of reaction and separation in a unit, directly leads to the introduction of one (or more) phases, or new reaction(s) in the system. For example, if one introduces a distillation operation in a liquid-phase catalytic reactor, vapor phase will be added to the system. Similarly, the adsorption could be combined with a catalytic equilibrium reaction in situ, where the new flowing solids phase is added to the classical packed bed reactor. Another well-known example is reactive absorption, where reaction is introduced in the liquid phase in order to enhance mass transfer and therefore the overall separation process. Thermodynamic number of degrees of freedom (F) can be expressed by the Gibbs' phase rule, as follows:

$$F = 2 + n_{\text{components}} - n_{\text{phases}} - n_{\text{reactions}} \quad (1)$$

where $n_{\text{components}}$, n_{phases} and $n_{\text{reactions}}$ are number of components, phases and reactions in the system, respectively. From Eq. (1) it is evident that the introduction of either a phase or a reaction lowers the number of degrees of freedom.

Schembecker and Tlatlik [59] have pointed out the reduction of degrees of freedom using an illustrative example which includes chemical reaction and membrane separation, represented in Fig. 3.

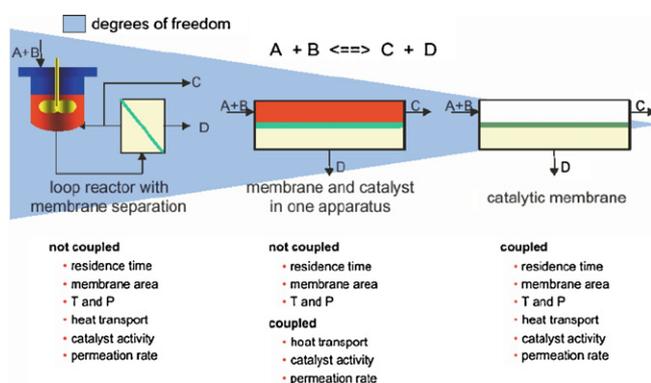


Fig. 3. Reduction of degrees of freedom in multifunctional reaction–membrane separation system.

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When a traditional stirred-tank reactor with separate membrane module and recycle is used, most of the variables are not coupled (see left side of Fig. 3), which allows more design and control options. Disadvantages of this consecutive approach arise from low product yield due to equilibrium limitations and high investment costs for two separate units. If reaction and separation are performed in one unit, reduction in capital cost is apparent. In this set-up energy savings could be achieved by heat integration, i.e. via direct use of reaction heat. However, the system has less DOF (see central part of Fig. 3) as the new phase (membrane) is introduced to the reactor and therefore some variables have become dependent. In a fully integrated system, where the catalytic membrane is used, removing of one product in situ leads to higher product yield (chemical equilibrium shift). This has positive effect on dimensions of the hybrid unit, as well. Nevertheless, in this case, most of the variables are coupled and the DOF are further reduced (right side of Fig. 3).

In brief, when process units are divided, there are more possibilities for measurement and manipulation of streams in between units. Furthermore, it is possible to install buffers and sinks in between separate units in order to effectively smooth the disturbances, which is not the case for integrated units. Moreover, due to more tight dependence between process' variables in the integrated process, independent control loops may not be adequate. Overall, process disturbances are likely to affect the whole integrated process and hardly could be localized by a single-loop control system [60].

Revealed difficulties for control of multifunctional units give a strong emphasis to a careful examination of actuation possibilities. This eventually could lead to exploration of novel process actuators. For example control of catalyst-membrane temperature via conductive electrical heating or microwaves' heating could improve performance of the hybrid reactor. The other possibility is to use alternative operation policies, like unsteady-state operation or time-varying feeding of reactants. Moreover, DOF reduction related problems could be further minimized if a mutual optimization of process design and control is performed at the early stage of conceptual design. For example if the shape and size of a catalytic membrane is optimized to suit both process requirements and controlled heating.

Combination of several functions in one unit has also restrictive effects in operation of a hybrid system. Every unit operation (e.g. reaction or separation) has its own feasible domain regarding process variables such as concentration, temperature, pressure. The physical and chemical limits for a certain operation border the so-called operation window. There is also a window for realistic design of units, which come from construction limitations of

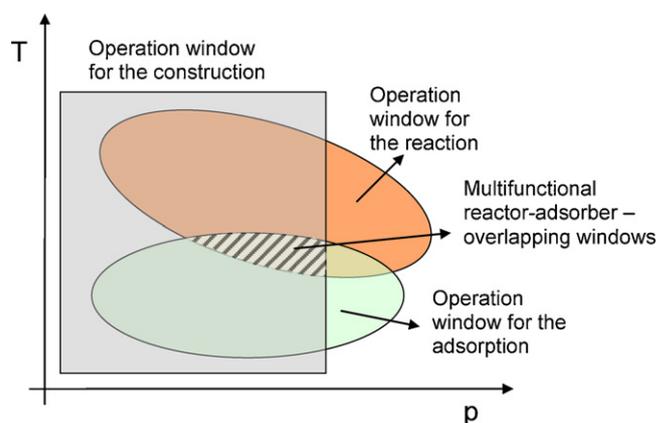


Fig. 4. Superposition of the operation windows (temperature and pressure) for the multifunctional reactor–adsorber.

materials. In classical units, one can employ any value of variable which allows feasible equipment design and it is usually chosen according to economical (and environmental) criteria. In multifunctional units, however, the windows for various operations are superimposed and one has to choose the operating point in the overlapping area. For example, operating windows of a reactor–adsorber are presented in Fig. 4. As it can be seen in Fig. 4, the chemical reaction requires higher temperatures for sufficient reaction rates. On the other hand, the adsorption has to be realized under lower temperatures and higher pressures. Too high pressures are not practical from the construction point of view. All together, the operation of a hybrid is restricted to a narrow area of overlapping windows. This simple analysis provides the essential information about potential feasibility of a multifunctional system, and should be performed before any conceptual design. It also offers information for integrated optimization providing the constraints for variables.

To summarize, the analyzed characteristics of functionally integrated systems may result in the following operation and control issues:

- (1) Reduction in number of degrees-of-freedom directly relates to decrease of actuation options (variables).
- (2) Disturbances cannot be easily smoothed due to the system integration.
- (3) More interactions between process variables contribute to non-linear behavior and add complexity to (model based) control.
- (4) Narrower operating windows limit actuation variables' domain.

4.2. Alternative driving forces and energy sources—dynamic advantages

The use of alternative driving forces and energy sources (*thermodynamic domain* of PI [5]) exhibit a large potential for process enhancement, concerning both the chemical reaction and the mass or heat transfer rates. Examples of different types of driving forces and energy fields reported in the literature include [61,62]:

1. Hi-gravity – generated by centrifugal forces for heat and mass transfer enhancement [10,12,63–67].
2. Electric – for droplet size manipulation and intensive mixing in liquid-liquid extraction [68].
3. Electromagnetic, microwave – for acceleration of reaction rates in organic synthesis [69].
4. Electromagnetic, light – for significant improvement of reaction selectivity [70].

5. Acoustic, ultrasound – for speeding up reaction rates and increase of product yield [71].

Most of the listed alternative energy sources have a common characteristic—a fast inherent dynamics. For example, electric and electromagnetic fields like microwaves or radio-frequency waves could be used for heating. For some liquids this heating is faster than a conventional heating via external vessel wall (or inserted coil). For some solids, the difference in heating dynamics between classical and electro(magnetic) is even more evident.

Fig. 5 displays the comparison of heating dynamics and temperature field uniformities, between the classical system with external wall heating and the alternative microwave (MW) heating. The system, in both cases comprises laminar flow of ethyl glycol in a narrow tube (dia. 5 mm, length 200 mm). The inlet superficial velocity is 0.1 m/s. In the case of conventional heating, constant heat flux is employed through the whole boundary (surface perpendicular to the fluid flow). The heat capacity of the metal wall is neglected. The overall external heat inflow is 165 W, which corresponded to a heat flux of 52.52 KW/m². For the microwave heating case, special attention is paid to the design of the applicator in order to attain homogeneous electromagnetic field and consequently uniform dielectric heating at the exposed part of the tube [72]. The dimensions of the rectangular cavity are 43 mm × 86 mm × 200 mm, and the exposed length of the tube is 43 mm (the middle part of the tube, indicated by the lines in Fig. 5b). In order to obtain high efficiency of the absorbed energy to the one reflected from the applicator, the dimensions of the wave excitation port and distance from the fluid tube have been adjusted. In this manner, the 94% utilization efficiency is achieved. Port power level is 300 W. The effects of glass tube wall were neglected. The 3D simulation has been performed in COMSOL MULTIPHYSICS, with a transverse electric (TE₁₀) electromagnetic propagation mode.

Fig. 5b shows 2D temperature fields (in the part of the tube) for microwave heating (left plot) and surface heating (right plot). The plots at 10 s are presented which corresponds to (nearly)

steady state for both cases. It is apparent that microwave heating resulted in more uniform radial temperature distribution in comparison to surface heating. This outcome could be expected since microwave heating is volumetric, while surface heating is localized to a boundary, resulting in high temperature gradients. Of course, the temperature field for microwave heating is not fully uniform—higher temperatures of ethyl glycol are to be located closer to the wall due to the radial distribution of velocities in laminar flow. It should be noted that uniform electromagnetic fields in fluids are not always easily achievable and an occurrence of hot spots could be common. In that sense, a special attention should be paid to the design and construction of microwave applicators, as mentioned before in the text and studied in [72].

Fig. 5a presents comparison of the heating dynamics, for microwave and surface cases. It is visible that microwave heating is somewhat faster, with lower time constant. The difference in heating speed in favor of microwaves would be more pronounced for lower surface to volume ratio of the system. Lower specific surface area would cause slower heating of an overall volume in case of classical heating, while this would not affect volumetric MW heating dynamics.

In the previous example one could notice that surface heating is more efficient in terms of energy consumption—165 W in contrast to 300 W for microwave heating for reaching the same average outlet temperature. This rather large discrepancy comes also from the high surface to volume ratio of the selected tube (small diameter). Furthermore, microwave heating is concentrated in 21.5% of the total tube volume. So again, if the tube diameter is larger and the tube length is shorter (lower surface to volume ratio), energy consumption efficiency of microwave heating would be improved. However, for achieving acceptable microwave heating uniformity, the diameter of the tube is constrained by the wavelength and dimensions of a cavity. Larger tube diameters correspond to large cavities and higher wavelengths, which is quite restrictive for commercial use [72]. Consequently, for industrial heating applications, the use of electromagnetic waves of lower frequencies (than usual

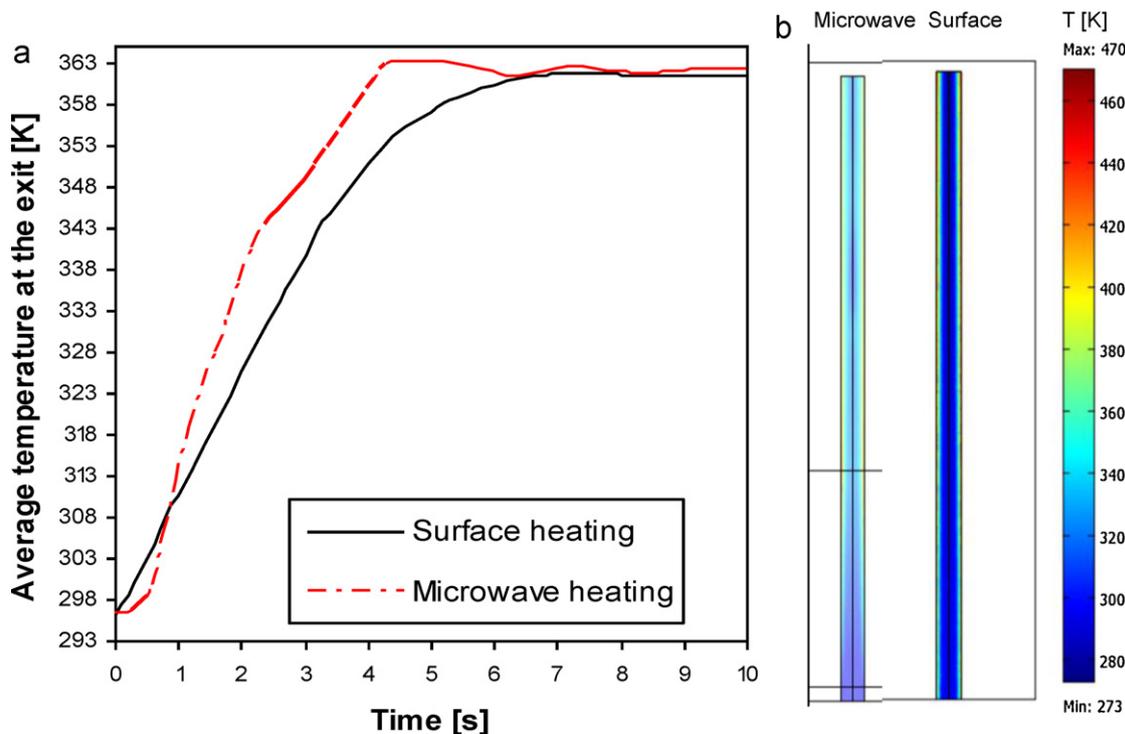


Fig. 5. Comparison between microwave heating and wall surface heating of the ethyl glycol flowing through a narrow tube: (a) heating dynamics, (b) temperature field uniformity (left – microwave heating; right – surface heating).

2.45 GHz) from the ISM band, like radio-frequency waves, probably would be more effective.

The difference in dynamic behavior between conventional heating and electro(magnetic) heating is caused by the difference in conductive and volumetric heating. That difference is even more evident for heating some solid materials. For instance, electro-resistance heating of solid electro-conductive materials, like equipment walls or porous monolith catalysis structures, could be effectively adjusted almost instantaneously, by tuning the power source. Similarly could stand for microwaves. This offers opportunities for efficient optimal temperature (heating) control of process systems.

The former arguments could be generalized for nearly all alternative driving forces and energy sources, including electrical, electromagnetic or even acoustic fields, for a number of reasons. Firstly, they are spreading almost instantaneously in space (with low energy losses) in comparison to much slower process transfer rates. Secondly, electrically based processes have short transient regimes and therefore their response to a change is very fast. In addition, they can be easily manipulated, by the change of electrical potential (or electrical current), in already well established technical solutions. These arguments could be extended to high gravity fields, which could be efficiently actuated for optimization of transfer fluxes. For instance, the rotational speed of spinning discs can be manipulated for control of mass transfer rates. In the case of multiphase reactions performed in spinning discs, gas-liquid interface area and mass transfer coefficient could be adjusted by rotational speed, as a bubble size increases significantly with a speed of rotation [73].

For above mentioned reasons, manipulation of alternative energy fields is a promising control option and possible novel (additional) actuator in intensified process system. However, a drawback for control could be still relatively sluggish response of fluid properties, i.e. change of concentration or temperature on a variation of electro(magnetic) fields. More important issue of using microwaves or electric current for temperature control is that cooling, if it is a required action cannot be employed with the same actuation system (e.g. microwave applicator). Furthermore, the fundamental (or empirical) mathematical dynamic relations between process variables and field intensity are not comprehensively developed. These models have to be derived and validated in order to be able to create efficient advanced model based control.

Alternative driving forces and energy fields can effect operation and control of intensified process systems, as summarized:

- (1) Due to their fast dynamics and ease of manipulation, alternative fields can be effectively exploited as new actuation variables.
- (2) Addition of phenomena in a system increases the complexity of a model to be used for control and optimization.
- (3) Effects of utilization of new phenomena in processes are not yet completely understood, thus dynamic behavior is hard to accurately predict.

4.3. Miniaturization of space—reduction in time

Miniaturization is one of the well recognized PI methods which falls in the *spatial domain* of PI [5]. Micro- and milli-systems have high surface to volume ratio, and therefore they directly contribute to the one of the PI principles, allowing high interfacial heat and mass transfer fluxes. Table 3 presents a simple illustration—the comparison of specific heat exchange surface areas, pressure drops and heat transfer coefficients for water flowing through the pipes of different sizes, varying from large-scale to micro-scale. In the example it is assumed that water with the average temperature of 50 °C is cooled by the wall with constant temperature of 40 °C. The flow capacity is identical for all systems ($Q=0.02 \text{ m}^3/\text{s}$) and the

Table 3

Comparison of heat transfer fluid-to-wall performance between pipes of four orders reduction in diameter.

	1	2	3	4
d (mm)	500	50	5	0.5
N	1	100	10,000	1,000,000
A (m^2/m^3)	2.5	25.5	254.6	2546.5
Re	92,000	9200	920	92
$\Delta P/L$ (mbar/m)	0.0019	0.0333	0.7132	71.317
Nu	533.4	84.5	4.10	3.67
h ($\text{KW}/\text{m}^2/\text{K}$)	0.619	0.981	0.475	4.257
hA ($\text{KW}/\text{m}^3/\text{K}$)	1.6	25.0	121.0	10,841.1

superficial velocities remain unchanged which implies increase in the number of pipes (N) with decreasing pipe diameter. It is assumed that water flow is distributed equally through all pipes. The heat transfer coefficient is calculated by well-known empirical correlations [74] for the Nusselt number (Nu). The flow in the smallest channel (0.5 mm) falls into the continuum flow with no-slip boundary conditions.

The drastic increase in hA value for micro-channels, with nearly 10,000-fold raise with respect to the largest pipe, is evident. This straightforward example illustrates the overall potential of micro-systems for efficient temperature control and disturbance rejection. However, since heat removal/supply from reaction medium in microreactor is very fast, conduction of heat through construction material may become dominant contribution and determine the time constant. Therefore, a wall material properties and design options (mass of construction material) become important for control.

Evidently, even higher heat and mass transfer rates could be achieved in smaller micro-channels, with characteristic dimension down to 100 μm or less. On the other hand, Table 3 shows that for 500 μm dia. even million pipes have to be installed in order to reach the same capacity at the same average superficial velocity as in the large-scale pipe. Consequently, the costs for the micro-system are much higher than for the large-scale system—capital ones for a more complex construction and more construction material per reaction volume, and operational ones, due to significant increase of the pressure drop in the micro-channels (Table 3). Furthermore, a large number of parallel channels may cause problems in operation and control. First of all, fluid has to be distributed evenly in very large number of small channels which is still a design challenge. Moreover, narrow channels may also be subjected to fouling and eventually blocking which could cause serious operating problems. A reasonable control strategy is to control a cluster or a block of parallel channels instead of controlling individually each one. This implies sensing at the output of a cluster where the fluid flows from the channels mix. Similarly, the actuation is employed at the entrance, before the distributor, or alternatively, at some point along the channels. In this arrangement, the control is designed according to the assumption that the process proceeds identically in all channels. However, blocking of only a certain number of the channels leads to observability problem. Eventually this may result in inappropriate control action, applied evenly to all of the channels of a cluster. These possible operation and control issues add to the existing design challenges for micro-scale systems, confirming a strong argumentation in favor of using millimeter size systems for industrial purposes. Milli-systems apparently are less sensitive to blocking, and a fewer number of channels are easier to operate. Obviously, in milli-systems heat and mass transfer enhancement is not as prominent as in micro-systems as depicted in Table 3. In conclusion, these conflicting objectives lead to a potential optimization problem for a design and operation of miniaturized systems.

Significant reduction in volume or fluid holdup offer more advantages to add to the above discussed enhancement of

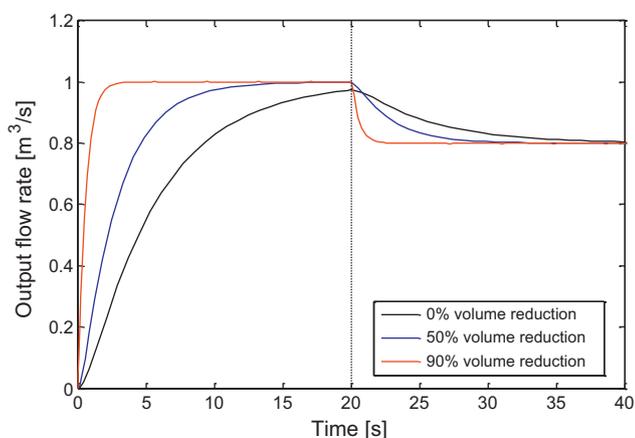


Fig. 6. Dynamic responses of the systems of different size on the step changes in feed flow rate. Outlet flow rate in time for: (a) base case volume (black line); (b) 50% volume reduction (blue line); (c) 90% volume reduction (red line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

transfer rates. Smaller volumes, for the same capacity, apparently lead to smaller investment costs, less energy consumed and inherently safer design due to less quantities of toxic material. From process dynamics point of view, smaller volumes for same flow rates produce faster process responses on the variables alteration. Consequently, smaller equipment is more sensitive to disturbances, which presents a challenge for control. As shown in the previous example, in micro-channel systems, disturbances could be annulled rapidly due to the high transfer rates. However, this potential has to be realized by the proper control system, which generally needs to be faster for volume-reduced systems. To illustrate this issue, Fig. 6 presents the dynamic responses of simple tanks-in-series system to a step change in input flow rate. The non-reactive system consists of two interconnected level tanks (of same size) with (level proportional) valves at the outlet streams. It is assumed that means of process intensification have contributed to the volume (surface) reduction of 50% (b) and 90% (c) in comparison to the base case volume (a). The input feed rate was same for all cases ($1 \text{ m}^3/\text{s}$), and it is decreased for 20% at time 20 s. Fig. 6 displays the outlet flow rate responses for the three systems of different size, firstly for the start-up period (up to 20 s), followed by the response to the inlet feed rate drop.

Evidently, the reduction in the volume capacity produces faster response in the outlet stream, i.e. the process time constant is decreased. Fig. 6 clearly shows that the dynamic response for the smallest system is in the range of several seconds, which is the order of magnitude less than for the case with no volume reduction. This has to be related to control system dynamics in order to have a prompt action—fast rejection of undesired disturbances. Therefore, the control decision (controller) and the action (actuator and sensor dynamics) should be adjusted to this fast process dynamics. This could become a problem if process dynamics becomes too fast. This argument can be generalized for all very fast processes, like millisecond catalytic reaction systems or similar.

Luyben and Hendershot [75] have discussed possible disadvantages of intensified processes in a sense of reduction of holdups. They have analyzed control closed-loop dynamics, using Aspen Dynamics, for four process examples: (1) benzene–toluene–xylene separation in two distillation columns – reduction of tray holdups; (2) Benzene nitration reactor(s) – two smaller reactors vs. one much larger; (3) Nitration reactor – fed batch reactor vs. CSTR; (4) HCN/ H_2O distillation – reduction of liquid holdup. The authors conclude that controllability and disturbance rejection studies need to be performed in advance and those results have to be considered

when designing intensified (meaning volume-reduced) processes. They emphasize that smaller holdups give the operator and control systems less time to respond to disturbances, and even suspect that process could be moved into unsafe regions of operation [75]. However, we consider that these setbacks could be avoided via a proper design of advanced control system; though we agree that dynamic analysis should be performed from the very beginning and should be incorporated in the conceptual design phase.

Process intensification through size reduction also underlines the importance of the dynamics of other control system elements, like measuring devices and actuators. Reduced process responses times, in the order of seconds or less, are comparable, or sometimes even smaller, to sensors' and actuators' dynamics. Therefore, the dynamics of all control loop elements should be considered when designing a control structure or even before, when examining the dynamics of the intensified process.

Furthermore, efficient control of intensified processes in many cases will require development of completely new sensors, or considerable improvement of existing ones. The advanced sensors are needed to determine whether products are within specification and whether processes are running optimally. The sensors have to cope with fast process dynamics and advanced control demands, and current instrumentation is usually too slow, inaccurate and working off-line. The novel sensors have to be able to precisely measure rapid-varying process variables on-line in process systems which are usually either miniaturized or have complex structures. This is a challenging demand for both fundamental research and sensors development and manufacturing.

Optical analytical techniques, such as spectroscopy (UV, Raman, NIR, VIS) or refractive index detection, have good prospects of implementation due to their versatile usability in combination with 'non-invasive' sensing characteristics. The advantage of spectroscopy is that the measurements are taken using optical fibers, requiring only a small measuring dot, which can be applied in situ for online analysis. Microchips for signal transmitting and integration of optical analytics for small volumes have been developed to some extent, but further applied research is necessary [76]. Other promising examples of novel sensors–actuators are small microelectronic devices which are supposed to float inside certain process equipment [77]. Such sensors use the ultra-wide band technology for transmitting large amounts of data with very low power over relative short distances. The key aspect is the proper design of sensors with respect to size, density, robustness and fluid compatibility which would enable the spatial and temporal monitoring of process variables in the entire volume of equipment, without causing considerable flow disturbances. Furthermore, the actuation may also be added to those devices allowing the local control of different process variables. However, the development of 'smart floating' sensors is still in early phase, and it needs more fundamental research.

In conclusion, in *spatial* domain of PI, miniaturization may encompass the following consequences for operation and control:

- (1) Very high heat transfer rates enable potential for fast and efficient temperature control, which may become dominantly a control of wall temperature of the micro/milli-unit.
- (2) Blocking of a number of narrow channels may lead to sensing, actuation and control difficulties in micro-systems.
- (3) Reduced process timescales require much faster control reactions than in classical systems, which may become a control system issue.
- (4) For fast process systems, the dynamics of sensing and actuation elements need to be accounted for in control system design.
- (5) New, smaller and faster sensors and actuators may be needed.

5. Towards the full integration of process intensification, operation and control

The presented analysis has demonstrated that intensified process systems have specific characteristics which may cause difficulties in operation and control of intensified systems, even if advanced model-based control systems are used. In order to efficiently handle these challenges, it is useful to consider operation and control in the earliest phases of conceptual process design (process synthesis). Preferably, process design, operation and control should be considered simultaneously, or in other term, they should be fully integrated. For that purpose, we attempt to postulate a new concept for process synthesis, outlined further in the text. This approach would not only provide optimal and safe operation, but may suggest innovative process solutions which are more economically beneficial and more environmentally friendly.

The interface between process and control is defined by actuation and sensing. Therefore, a systematic analysis of actuation contributes significantly to the integration of process design, operation and control. Moreover, actuation analysis and design may provide additional options for handling mentioned operation and control issues of intensified process systems. This analysis entails a careful exploration of (intensified) processes fundamentals in the sense of possibilities for actuation. As a result, new actuators may arise, as illustrated in Section 4, which will be discussed more systematically in the following text.

5.1. Actuation analysis and design

Generally, actuation enables the manipulation of system behavior in the sense of control and optimization. So far, actuation has not received much of attention in the literature. A possible explanation is that control engineers consider it to be unchangeable result of process design, while process engineers regard it as fixed for a unit operation. Huesman et al. [78,79] argued that the systematic outlook on actuation, which leads to actuation design and its intensification, may result in considerable economical and control improvements.

In majority of chemical processes four physicochemical phenomena play an essential role: (a) chemical reaction, (b) mass transfer, (c) heat transfer and (d) momentum transfer. It should be noted that by applying process intensification concepts, more phenomena than those four can be dominant in a process, e.g. energy transferred by microwaves. However, to underline the main point, only main four will be considered. These phenomena are characterized by the fluxes (rates for reaction) and geometry (volume and surface). Therefore, actuation implies manipulation of fluxes and/or geometry. Fluxes occur as a result of deviations from thermodynamic equilibrium and can be expressed by multiplication of the driving force and the transfer coefficients, for example:

$$J_q = h\Delta T \quad (2)$$

where J_q is the heat flux, ΔT is the driving force–temperature difference and h is the heat transfer coefficient. Heat transfer is given by:

$$F_q = J_q A = h\Delta T A \quad (3)$$

Eq. (2) shows that heat transfer could be actuated by h , or/and ΔT or/and by or A . The heat transfer coefficient could be altered, for example, by the fluid velocity or stirring velocity. Temperature difference could be manipulated by the temperature of the heat medium or by the wall temperature. The surface area is a design parameter, which could be optimized also for a control (in simultaneous optimization). However, in most cases it is nearly impossible to manipulate the surface once the system is realized.

The presented analysis can be straightforward generalized for the other physicochemical phenomena which may be manipulated by: (a) coefficient, (b) driving force and (c) surface area or volume. For the purpose of actuation efficiency enhancement, process intensification methods could be exploited in many ways. For example, in the *functional domain*, a manipulation of an additional phase (like vapor in reactive distillation or particles in reactive adsorption) may alter the driving force for an equilibrium reaction.

In the *thermodynamic domain*, the use of alternative energy sources, like microwaves or electrically induced conductive heating, could improve manipulation of heat fluxes which was already addressed in Section 4 of the article. In another example from that domain, the change of rotation speed in rotating packed beds or spinning disc reactors could be used for alteration of mass transfer coefficients and interfacial areas.

In the *spatial domain*, above mentioned variable geometry designs could be ‘tailored’ for the optimal local transfer rates, for example applied to micro- and milli-systems. Furthermore, spatial actuation could be applied to spatially distributed systems, for example multiple input points for tubular reactors, microreactors, packed bed reactors, etc. Spatial actuation could be used to attain optimal concentration and/or temperature profiles and to improve controllability of such systems. The use of miniature sensors/actuators floating inside the vessels is another opportunity for spatial/local actuation of process systems, which was discussed in Section 4.

In the *temporal domain*, deliberate transient operation of processes could lead to improved actuation as it can cause the change of all three terms: coefficients, driving forces and even interface surfaces.

However, effects of these alternative actuations have to be examined in more depth theoretically and experimentally. Such analysis could show the realistic opportunities for intensification of actuation. Without doubt, some of those would turn out impractical from a realization point of view.

5.2. New integral approach to process synthesis based on dynamic optimization

Traditional process synthesis (PS), based on the general concept of unit operations, was established by Sirola and Rudd [80] and later extended by Douglas [81]. These concepts are based on systematic, but heuristic approach, relying for a part on the designer's experience. Although many other, more advanced, methods have been proposed, the basic PS concept is still dominant in practice. The underlying approach to process synthesis contains several consecutive stages, as depicted in Fig. 7a.

In order to overcome limitations of design based on series of predefined unit operations several authors introduced the concept of phenomena-driven PS [82–84]. A common background for these approaches is the use of process tasks or functions, related to process phenomena, to achieve certain process objectives. Although these concepts theoretically allow innovation and PI, relevant examples were not presented so far. Moreover, they did not use rigorous optimization and thus could undergo the same downsides as the traditional concepts described formerly.

The second largest class of PS methods use optimization, thus ideally offering optimal process solutions. Even though optimization techniques, both deterministic and stochastic, have advanced in recent times, there are still difficulties associated with optimization problem formulation and efficient program convergence [85]. More importantly, they are mostly based on unit operations, except for some contributions related to reactive distillation and hybrid separation systems [86–90]. Extensive review on conceptual process design methodologies and their implementation on different process scales is given by Li and Kraslawsky [91].

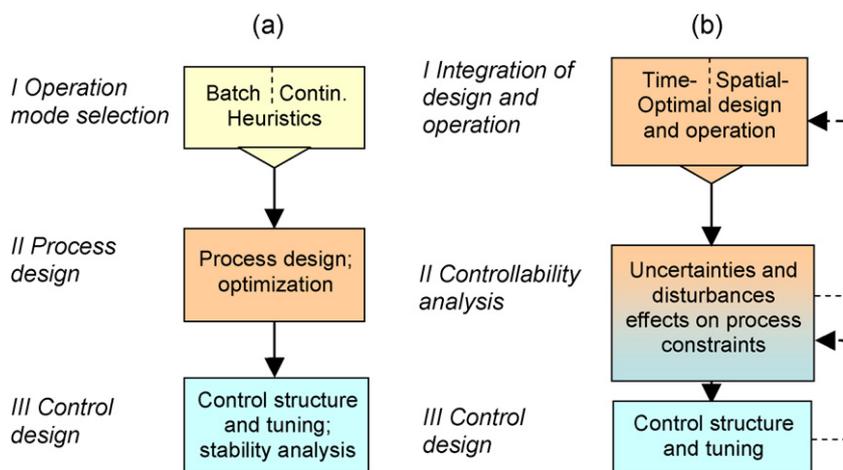


Fig. 7. Process synthesis schemes: (a) traditional approach based on heuristics and unit-operations concept; (b) new approach based on optimization and exploitation of process intensification concepts.

In their recent work Freund and Sundmacher [92,93] have managed to combine phenomena-driven approach with optimization, in a so-called concept of elementary process functions. However, their methodology is demonstrated only on a chemical reactor [93] with no extensive PI content, it does not include design-control interactions and does not account for uncertainties. Recently, Lutze et al. [94] have offered a systematic framework for process synthesis which examines different process intensification options. The authors [94] proposed a hierarchical sequence of steps, in order to gradually reduce PI options, ending up with feasible optimization. They also constructed a knowledge base to provide necessary information for intensified processes. Although promising, this concept is limited to continuous operation of existing intensified equipment (retrofitting) and also does not consider uncertainties or control and operation at all.

Interactions between process design and control have been studied broadly since 1990s and integration methodologies are mainly based on optimization [95–99]. A commonly used framework for simultaneous design-control dynamic optimization under uncertainty, has been introduced by Bansal et al. [99]. In several articles, interactions of design and control, for reactive distillation columns have been studied [100–102]. However, these contributions mainly considered classical control systems (PID) and they did not include diverse process intensification methods in the integrative PS framework. Further, actuation, which essentially determines the interface between design and control [78,79], was not analyzed thoroughly.

Up to date, process synthesis concepts of all kinds implied the selection of the operation mode as the first step of the conceptual process design (CPD). Several operation modes are very well established in process systems, namely: continuous, batch, fed-batch and periodic. The choice of particular mode is exclusively based on heuristics. For example, batch operation is recommended for lower production rates and for multi-product processes, due to its flexibility. Continuous operation is applied for bulk production (large capacities) of a single product. The selection is made based on traditional equipment and on rough economic estimates. However, it is reasonable to assume that process operation and its profitability is seriously limited by fixing the operation mode *a priori*. The reasons are grounded in the heuristics and very small number of possible options. Therefore, we suggest that operation mode should not be fixed at the beginning of CPD, yet it should be incorporated in simultaneous optimization problem. In that sense, the operation selection could be ideally seen as a decision problem with a continuous solution space.

We are suggesting a new concept for process synthesis, which integrates some of the previous concepts (mentioned above), attempts to overcome their insufficiencies and introduces novel approaches. This optimization-based concept is presented schematically in Fig. 7b, parallel to a traditional and well-established approach (Fig. 7a). Our concept contains also three (but merging) stages, though it is qualitatively different from classical one—it provides optimal process solution and allows strong interactions between process design, operation and control. In the first stage, the integration of process design and operation should be attained by means of dynamic optimization (optimal control), based on simple first principles mathematical models. Optimization objectives are based on economic factors and they also include environmental constraints and costs. Optimization results at this stage should provide optimal process design and optimal operation on a conceptual level with theoretically achievable efficiency.

The first phase of proposed process synthesis concept is determined by optimization of fluxes and driving forces within a process, thus it goes beyond the limits of the traditional unit operation approach. In order to achieve/approach these theoretically optimized process conditions, the concept exploits process intensification principles and combination of several PI methods in a process design. It also explores the possibilities for actuation improvement for optimal operation and control, as described above in the previous subsection.

In the second stage (see Fig. 7b) controllability study for the optimal process solutions is to be performed, by means of robust optimization techniques. In most cases, optimization in the first phase will clearly distinguish few options which markedly outperform the others. For these nominally optimal cases, the effects of model uncertainties, specifically physical parameters uncertainties, and process disturbances on the process constraints and overall performance are to be examined. This analysis may lead to an actuation adjustment in the operation domain, in order to avoid the violation of the constraints, which is presented by the dashed feedback line to the first stage in Fig. 7b. Eventually, the results of the analysis may even require some design alterations (phase one) in order to have more reliable process, which operates under uncertainties.

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 adjustments (dashed line to the first stage).

The first issue that could arise regarding the most important first stage of the suggested concept is how to effectively formulate a problem in a single mathematical model and optimization structure. Usually a very large number of options can be set for one process, and therefore a single optimization program would be very difficult to derive and solve. Certainly, some process options are unfeasible and could be discriminated at the beginning. Lutze et al. [94] proposed a systematic framework with a hierarchical sequence of steps and a base of PI models, which could be a tool for establishing solvable optimization structure. However, this methodology still includes heuristics and requires highly experienced process engineers and modelers. The problems could probably be tackled by a combination of proposed PS methodologies with tools for automatic generation of the dynamic process system models. These model-generation methods are currently in research focus of academic groups and companies [103,104]. For example, the initial problem set-up could be provided by a framework for a construction of models based on the network modeling, published recently by Preisig [103]. However, complete development of the mentioned concepts, their mutual integration in user-friendly computer environment should be seen as a longer term ambition.

The second potential challenge for the concept realization is the solving of the mixed-integer dynamic optimization with many variables for optimization, lot of different constraints and possibly multi-objective target function. However, the optimization techniques and partial-differential-algebraic equation (PDAE) solvers are in the mature state, and they will progress more [105,106]. Yet, optimization may limit the complexity of the mathematical models to be used in the concept. For that reason, at the present, only simplified models depicting relevant phenomena are to be formulated. Nevertheless, the results in the first stage should clearly offer optimal process design and optimal operation, demonstrating considerable cost reductions.

The challenges for the second stage would be to implement uncertainties in the optimization set-up, i.e. to formulate and run a robust dynamic optimization. Moreover, an adequate and efficient controllability criteria needs to be developed, or existing controllability criteria (output-input, state, functional, thermodynamic, etc.) have to be improved and adapted. For the third stage, which consists of the process control design, challenges and possibilities are reviewed in the previous sections.

The presented PS approach addresses both the meso- and macro- scale of chemical processes, as it operates with phenomena, fluxes and phases (meso-) as well as with apparatus, processes and plants (macro-). The next logical step would be to extend it in two directions, towards molecular scale on one hand and the mega-scale on the other. On the molecular scale it could be beneficial to include different chemical reaction routes to a desired product in one process optimization. That way more degrees of freedom could be provided for process synthesis—exploring more process options. For example, two chemical synthesis routes might differ in undesirable side products and consequently different separation methods could be examined for each route. In another case, different chemical routes might suggest different reactor types and conditions. For this integration, contribution from (green) chemistry and product design are highly valuable.

On the other pole, interactions between process synthesis and a company's management plans and supply chain management, can offer advantages in operation and consequently lead to higher profits. For a conceptual process design phase, various scheduling scenarios could be examined and optimized integrally with

process design and control. Certainly, the full economic potential could be attained in on-line and real-time integration of management actions with higher-level optimal control system. However, the overall integration of multi-scale approaches in optimization-based process synthesis is a challenging target on a long term.

To summarize, proposed synthesis concept focuses on providing theoretical limits of process' efficiency by exploiting various PI methods and actuation possibilities in a process, and using dynamic optimization to attain the integration of process operation, design and control. The concept is not restrained to current technical issues. Expected significant improvements in technological and economical performance of process solutions, obtained by the suggested concept, will give a motivation for their further development and practical realization.

6. Concluding remarks

The current societal requirements and the state of environment, call for accelerated development of more sophisticated and efficient process systems. Process intensification concepts have offered significant advantages in material and energy efficiency in diverse process applications, up to date mostly by using PI methods in design of novel types of equipment. Further advances in economical and environmental performance can be achieved if operation and control of a whole process are considered simultaneously and systematically together with different PI design options.

The dynamic characteristics of intensified process systems bring both challenges and opportunities for process operation and control. Multifunctional systems, in which process operations or functions are integrated, have less degrees-of-freedom than classical processes, which can limit control options. Moreover, the multifunctional units operate in narrower operating range (than separated unit operations), which also can be reflected in actuation restriction. On the other hand, alternative driving forces and energy sources can be exploited for control, as new actuation possibilities. For example, electric or electromagnetic fields can be effectively used for precise heating control of solids or fluids. This is due to the fast dynamics of energy propagation, possibility of selective heating, short transient regimes and ease of manipulation. Miniaturized systems also offer advantages for control, as they provide very high mass and heat transfer rates. For instance, any disturbance in temperature can potentially be suppressed rapidly, as the overall heat transfer resistance is very low in a micro-scale system. However, miniaturization directly relates to the large reduction in process timescales. This means that control decisions and actions need to be in accordance with the small process timescales; which in some cases implies faster control systems than existing ones, and thus may become an issue. Further, due to miniaturization, new class of sensors needs to be developed, to fit in a system.

After careful adjustment to the mentioned dynamic specifics, advanced control systems, realized in hierarchical layer-type structure, will be capable of controlling and optimizing intensified process systems. Apparently, model based control and optimization is the essence of such systems. Still an open issue is a fast and reliable modeling of intensified process systems customized for on-line control applications (which include optimization). Additionally, some advances should be made for efficient implementation of non-linear MPC, as the models of intensified systems are highly non-linear in most cases.

Although many process synthesis concepts have been proposed in the literature, yet there is no an effective method to combine phenomena-based synthesis (process intensification fundamentals) with rigorous optimization tools. Such method would be beneficial economically and would promote process innovation. A

new approach to process synthesis, suggested in this paper, enables gradual integration of process operation, design and control, by means of dynamic optimization. It is based on utilization of process intensification concepts and their combination in one process. One of the key features of the suggested synthesis concept is a systematic exploration and design of actuation. The proposed process synthesis method firstly determines (simultaneously) the optimal operation and design of a process. Then uncertainties are considered through controllability analysis. Finally, taking into account the controllability results, optimal control is conceptually designed, which target is to follow optimal operation paths as effectively and robustly as possible.

The aim of the presented process synthesis is to provide theoretical limits of possible enhancement in terms of economic and environmental measures. If significant cost reductions are displayed as a result of the process synthesis, this could be a drive for companies to resolve existing technical issues and implement new process solutions in the industry. These new process systems would be smaller, more modular and standardized. They would be more agile, operating more often in dynamic regimes for which sophisticated and robust control systems would be created.

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References

- M.P. Dudukovic, *Frontiers in reactor engineering*, Science 325 (2009) 698–701.
- European Roadmap for Process Intensification, *Creative Energy–Energy Transition*. www.creative-energy.org, SenterNovem, 2008.
- J.A. Moulijn, A. Stankiewicz, J. Grievink, A. Górák, *Process intensification and process systems engineering: a friendly symbiosis*, *Comput. Chem. Eng.* 32 (1–2) (2008) 3–11.
- K.-U. Klatt, W. Marquardt, *Perspectives for process systems engineering—personal views from academia and industry*, *Comput. Chem. Eng.* 33 (3) (2009) 536–550.
- T. Van Gerven, A. Stankiewicz, *Structure, energy, synergy, time—the fundamentals of process intensification*, *Ind. Eng. Chem. Res.* 48 (5) (2009) 2465–2474.
- C. Ramshaw, *Higee distillation—an example of process intensification*, *Chem. Eng. (London)* 389 (1983) 13–14.
- W.T. Cross, C. Ramshaw, *Process intensification – laminar-flow heat-transfer*, *Chem. Eng. Res. Des.* 64 (1986) 293–301.
- A. Stankiewicz, J.A. Moulijn, *Process intensification: transforming chemical engineering*, *Chem. Eng. Prog.* 96 (1) (2000) 22–34.
- C. Tsouris, J.V. Porcelli, *Process intensification—has its time finally come?* *Chem. Eng. Prog.* 99 (10) (2003) 50–55.
- A. Stankiewicz, J. Moulijn (Eds.), *Re-Engineering the Chemical Processing Plant: Process Intensification*, Marcel Dekker, New York, 2004.
- F.J. Keil (Ed.), *Modeling of Process Intensification*, Wiley-VCH, 2007.
- D.A. Reay, C. Ramshaw, A.P. Harvey (Eds.), *Process Intensification, Engineering for Efficiency, Sustainability and Flexibility*, Butterworth-Heinemann, Oxford, 2008.
- V. Hessel, S. Hardt, H. Lowe, *Chemical Micro Process Engineering. Fundamentals Modelling and Reactions*, Wiley-VCH, 2004.
- A. Cubulski, J.A. Moulijn (Eds.), *Structured Catalysts and Reactors*, 2nd ed., Taylor & Francis, 2006.
- K. Sundmacher, A. Kienle, *Reactive Distillation Status and Future Directions*, Wiley-VCH, Weinheim, 2003.
- S. Becht, R. Franke, A. Geißelmann, H. Hahn, *An industrial view of process intensification*, *Chem. Eng. Process.: Process Intensif.* 48 (1) (2009) 329–332.
- G.J. Harmsen, *Reactive distillation: the front-runner of industrial process intensification. A full review of commercial applications, research, scale-up, design and operation*, *Chem. Eng. Process.: Process Intensif.* 46 (9 (SPEC. ISS.)) (2007) 774–780.
- H.G. Schoenmakers, B. Bessling, *Reactive and catalytic distillation from an industrial perspective*, *Chem. Eng. Process.* 42 (3) (2003) 145–155.
- T.F. Edgar, *Control and operations: when does controllability equal profitability?* *Comput. Chem. Eng.* 29 (2004) 41–49.
- P.S. Buckley, *Techniques of Process Control*, John Wiley & Sons, New York, 1964.
- W.L. Luyben, *Steady-state energy conservation aspects of distillation column control design*, *Ind. Eng. Chem. Fundam.* 14 (1975) 321–325.
- J.J. Downs, J.E. Doss, *A view from North American industry*, in: Y. Arkun, W.H. Ray (Eds.), *Chemical Process Control*, vol. IV, CACHE/AICHE, Austin, 1991, pp. 53–77.
- C.R. Cutler, B.L. Ramaker, *Dynamic matrix control—a computer control algorithm*, in: *Proceedings of AIChE 86th National Meeting*, Houston, TX, 1979.
- J.A. Richalet, A. Rault, J.D. Testud, J. Papon, *Model predictive heuristic control: applications to industrial processes*, *Automatica* 14 (1978) 413–428.
- T.E. Marlin, A.N. Hrymak, *Real-time operations optimization of continuous processes*, *Chem. Process Control vol. V, AIChE Sympos. Ser.* 93 (316) (1997) 156–164.
- J.V. Kadam, W. Marquardt, *Integration of economical optimization and control for intentionally transient process operation*, in: R. Findeisen, F. Allgöwer, L.T. Biegler (Eds.), *Assessment and Future Directions of Nonlinear Model Predictive Control*, Springer, Berlin, 2007.
- S. Engell, *Feedback control for optimal process operation*, *J. Process Control* 17 (3) (2007) 203–219.
- P.A. Rolandi, J.A. Romagnoli, *Framework for on-line full optimising control of chemical processes*, *European Symposium on Computer-Aided Process Engineering 15 (20A–B)* (2005) 1315–1320.
- A.E.M. Huesman, O.H. Bosgra, P.M.J. Van den Hof, M.P.C. Integrating, *RTO in the process industry by economic dynamic lexicographic optimization: an open-loop exploration*, in: *AIChE Annual Meeting*, Philadelphia, USA, 2008.
- D.E. Seborg, T.F. Edgar, D.A. Mellichamp, *Process Dynamics and Control*, Wiley, New York, 2004.
- D. Bonvin, B. Srinivasan, D. Ruppen, *Dynamic optimization in the batch chemical industry*, *Chem. Process Control – CPC VI, AIChE Sympos. Ser.* 97 (326) (2002) 255–273.
- M. Darby, M. Harmse, M. Nikolaou, *MPC: current practice and challenges*, *Proc. IFAC ADCHEM* (2009) (12–15 July, Istanbul, Turkey).
- L. Ljung, *System Identification—Theory for the User*, 2nd ed., Prentice Hall, Englewood Cliffs, New Jersey, 1999.
- Y. Zhu, *Multivariable System Identification for Process Control*, Elsevier Science Ltd., Oxford, 2001.
- N. Sharma, K. Singh, *Control of reactive distillation column: a review*, *Int. J. Chem. React. Eng.* 8 (2010) R5.
- M. Al-Arfaj, W.L. Luyben, *Comparison of alternative control structures for an ideal two-product reactive distillation column*, *Ind. Eng. Chem. Res.* 39 (9) (2000) 3298–3307.
- M.A. Al-Arfaj, W.L. Luyben, *Comparative control study of ideal and methyl acetate reactive distillation*, *Chem. Eng. Sci.* 57 (24) (2002) 5039–5050.
- M.G. Sneesby, M.O. Tadé, R. Datta, T.N. Smith, *Two-point control of a reactive distillation column for composition and conversion*, *J. Process Control* 9 (1) (1999) 19–31.
- D.A. Bartlett, O.M. Wahnschafft, *Dynamic Simulation and Control Strategy Evaluation for MTBE Reactive Distillation*, Presented at FOCAP, 1999.
- Y. Tian, F. Zhao, B.H. Bisowarno, M.O. Tadé, *Pattern-based predictive control for ETBE reactive distillation*, *J. Process Control* 13 (1) (2003) 57–67.
- B.H. Bisowarno, Y. Tian, M.O. Tadé, *Model gain scheduling control of an ethyl tert-butyl ether reactive distillation column*, *Ind. Eng. Chem. Res.* 42 (15) (2003) 3584–3591.
- R. Khaledi, B.R. Young, *Modeling and model predictive control of composition and conversion in an ETBE reactive distillation column*, *Ind. Eng. Chem. Res.* 44 (9) (2005) 3134–3145.
- A. Kumar, P. Daoutidis, *Modeling analysis and control of ethylene glycol reactive distillation column*, *AIChE J.* 45 (1) (1999) 51–68.
- N. Vora, P. Daoutidis, *Dynamics and control of an ethyl acetate reactive distillation column*, *Ind. Eng. Chem. Res.* 40 (3) (2001) 833–849.
- S. Grüner, K. Mohl, A. Kienle, E.D. Gilles, G. Fernholz, M. Friedrich, *Nonlinear control of a reactive distillation column*, *Control Eng. Pract.* 11 (8) (2003) 915–925.
- F.J. Doyle, L.S. Balasubramhanya, *Nonlinear model based control of a batch reactive distillation column*, *J. Process Control* 10 (2000) 209–280.
- M. Volker, C. Sonntag, S. Engell, *Control of integrated processes: a case study on reactive distillation in a medium-scale pilot plant*, *Control Eng. Pract.* 15 (7) (2007) 863–881.
- I.J. Halvorsen, S. Skogestad, *Optimal operation of petlyuk distillation: steady-state behavior*, *J. Process Control* 9 (1999) 407.
- J.G. Segovia-Hernández, S. Hernández, R. Femat, A. Jiménez, *Control of thermally coupled distillation sequences with dynamic estimation of load disturbances*, *Ind. Eng. Chem. Res.* 46 (2007) 546.
- M. Serra, A. Espuña, L. Puigjaner, *Control, optimization of the divided wall column*, *Chem. Eng. Process.* 38 (1999) 549.
- T. Adrian, H. Schoenmakers, M. Boll, *Model predictive control of integrated unit operations: control of a divided wall column*, *Chem. Eng. Process.: Process Intensif.* 43 (3) (2004) 347–355.
- F. Lode, M. Houmard, C. Migliorini, M. Mazzotti, M. Morbidelli, *Continuous reactive chromatography*, *Chem. Eng. Sci.* 56 (2) (2001) 269–291.
- A. Tounsi, S. Engell, *Optimization-based control of a reactive simulated moving bed process for glucose isomerization*, *Chem. Eng. Sci.* 59 (18) (2004) 3777–3792.
- V.M. Zahn, C. Yi, A. Seidel-Morgenstern, *Analysis and demonstration of a control concept for a heat integrated simulated moving bed reactor*, *Chem. Eng. Sci.* 66 (20) (2011) 4901–4912.
- S. Hasebe, *Design and operation of micro-chemical plants—bridging the gap between nano, micro and macro technologies*, *Comput. Chem. Eng.* 29 (1) (2004) 57–64.

- [56] O. Tonomura, S. Nagahara, J. Kano, M. Kano, S. Hasebe, Sensor location for effective fault diagnosis in micro chemical processes, in: Paper Presented at the IFAC Proceedings Volumes (IFAC-PapersOnline) 7(PART 1), 2009, pp. 309–314.
- [57] M.V. Kothare, Dynamics and control of integrated microchemical systems with application to micro-scale fuel processing, *Comput. Chem. Eng.* 30 (10–12) (2006) 1725–1734.
- [58] L.G. Bleris, M.V. Kothare, Reduced order distributed boundary control of thermal transients in microsystems, *IEEE Trans. Control Syst. Technol.* 13 (6) (2005) 853–867.
- [59] G. Schembecker, S. Tlatlik, Process synthesis for reactive separations, *Chem. Eng. Process.* 42 (3) (2003) 179–189.
- [60] S. Engell, G. Fernholz, Control of a reactive separation process, *Chem. Eng. Process.* 42 (3) (2003) 201–210.
- [61] A. Stankiewicz, Energy matters: alternative sources and forms of energy for intensification of chemical and biochemical processes, *Chem. Eng. Res. Des.* 84 (A7) (2006) 511–521 (Trans. IChemE, Part A).
- [62] A. Stankiewicz, On the applications of alternative energy forms and transfer mechanisms in microprocessing systems, *Ind. Eng. Chem. Res.* 46 (12) (2007) 4232–4235.
- [63] C. Ramshaw, Opportunities for exploiting centrifugal fields, *Heat Recovery Syst. CHP* 13 (6) (1993) 493–513.
- [64] A. Aoune, C. Ramshaw, Process intensification: heat and mass transfer characteristics of liquid film on rotating discs, *Int. J. Heat Mass Trans.* 42 (1999) 2543–2556.
- [65] C. Zheng, K. Guo, Y. Song, X. Zhou, D. Ai, Industrial practice of higravitec in water deaeration, in: J. Semel (Ed.), 2nd International Conference on Process Intensification in Practice, BHR Group, London, 1997, pp. 273–287.
- [66] D. Trent, D. Tirtowidjojo, Commercial operation of a rotating packed bed (RPB) and other applications of RPB technology, in: M. Gough (Ed.), 4th International Conference on Process Intensification in Practice, BHR Group, London, 2001, pp. 11–19.
- [67] M. Vicevic, K.V.K. Boodhoo, K. Scott, Catalytic isomerisation of α -pinene oxide to campholenic aldehyde using silica-supported zinc triflate catalysts. II. Performance of immobilized catalysts in a continuous spinning disc reactor, *Chem. Eng. J.* 133 (2007) 43–57.
- [68] L.R. Weatherley, Electrically enhanced extraction, in: J.J. Thornton (Ed.), *Science and Practice of Liquid–Liquid Extraction*, Oxford University Press, UK, 1992.
- [69] M. Larhed, C. Moberg, A. Hallberg, Microwave-accelerated homogeneous catalysis in organic chemistry, *Acc. Chem. Res.* 35 (9) (2002) 717–727.
- [70] T. Van Gerven, G. Mul, J. Moulijn, A. Stankiewicz, A review of intensification of photocatalytic processes, *Chem. Eng. Process.: Process Intensif.* 46 (9 (SPEC. ISS.)) (2007) 781–789.
- [71] L.H. Thompson, L.K. Doraiswamy, Sonochemistry: science and engineering, *Ind. Eng. Chem. Res.* 38 (4) (1999) 1215–1249.
- [72] G.S.J. Sturm, G.D. Stefanidis, M.D. Verweij, T.D.T. Van Gerven, A.I. Stankiewicz, Design principles of microwave applicators for small-scale process equipment, *Chem. Eng. Process.: Process Intensif.* 49 (9) (2010) 912–922.
- [73] M. Meeuwse, J. van der Schaaf, B.F.M. Kuster, J.C. Schouten, Gas–liquid mass transfer in a rotor–stator spinning disc reactor, *Chem. Eng. Sci.* 65 (1) (2010) 466–471.
- [74] J.L. Plawsky, *Transport Phenomena Fundamentals*, Marcel Dekker Inc., 2001.
- [75] W.L. Luyben, D.C. Hendershot, Dynamic disadvantages of intensification in inherently safer process design, *Ind. Eng. Chem. Res.* 43 (2) (2004) 384–396.
- [76] Action Plan Process Intensification, Creative Energy–Energy Transition, www.creative-energy.org, SenterNovem, 2008.
- [77] M. Motegh, Application of smart moving process environment actuators and sensors (smart peas) in process industry, OSPT Process Technology Miniposter Book, 2009, p. 72.
- [78] A.E.M. Huesman, P.M.J. Van den Hof, A.I. Stankiewicz, On the essential role of process control in process intensification, in: Proceedings of the 2nd European Process Intensification Conference–EPIC, Venice, 2009.
- [79] A.E.M. Huesman, O.H. Bosgra, P.M.J. Van den Hof, Towards the systematic of actuation for process systems, in: Proceedings of 9th IFAC International Symposium on Dynamics and Control of Process Systems, DYCOPS, Leuven, 2010.
- [80] J.J. Sirola, D.F. Rudd, Computer-aided synthesis of chemical process designs—from reaction path data to the process task network, *Ind. Eng. Chem. Fundam.* 10 (3) (1971) 353–362.
- [81] J.M. Douglas, *Conceptual Design of Chemical Processes*, McGraw-Hill, New York, 1988.
- [82] J. Tanskanen, V.J. Pohjola, K.M. Lien, Phenomenon driven process design methodology: focus on reactive distillation, *Comput. Chem. Eng.* 19 (Suppl. 1) (1995) 77–82.
- [83] I.S. Gavrilu, P. Iedema, Phenomena-driven process design, a knowledge-based approach, *Comput. Chem. Eng.* 20 (Suppl. 1) (1996) 103–108.
- [84] J.A. Arizmendi-Sánchez, P.N. Sharratt, Phenomena-based modularisation of chemical process models to approach intensive options, *Chem. Eng. J.* 135 (1–2) (2008) 83–94.
- [85] I.E. Grossmann, M.M. Daichendt, New trends in optimization-based approaches to process synthesis, *Comput. Chem. Eng.* 20 (6–7) (1996) 665–683.
- [86] W. Marquardt, S. Kossack, K. Kraemer, A framework for the systematic design of hybrid separation processes, *Chin. J. Chem. Eng.* 16 (3) (2008) 333–342.
- [87] P. Linke, A. Kokossis, On the robust application of stochastic optimization technology for the synthesis of reaction/separation systems, *Comput. Chem. Eng.* 27 (5) (2003) 733–758.
- [88] R.M. Dragomir, M. Jobson, Conceptual design of single-feed kinetically controlled reactive distillation columns, *Chem. Eng. Sci.* 60 (18) (2005) 5049–5068.
- [89] N. Dalaoui, P. Seferlis, A unified modeling framework for the optimal design and dynamic simulation of staged reactive separation processes, *Comput. Chem. Eng.* 30 (8) (2006) 1264–1277.
- [90] M. Urseilmann, S. Engell, Optimization-based design of reactive distillation columns using a memetic algorithm, *Comput. Chem. Eng.* 28 (2010) 1243–1248.
- [91] X. Li, A. Kraslawski, Conceptual process synthesis: past and current trends, *Chem. Eng. Process.: Process Intensif.* 43 (5) (2004) 589–600.
- [92] H. Freund, K. Sundmacher, Towards a methodology for the systematic analysis and design of efficient chemical processes. Part 1. From unit operations to elementary process functions, *Chem. Eng. Process.: Process Intensif.* 47 (12) (2008) 2051–2060.
- [93] A. Peschel, H. Freund, K. Sundmacher, Methodology for the design of optimal chemical reactors based on the concept of elementary process functions, *Ind. Eng. Chem. Res.* 49 (21) (2010) 10535–10548.
- [94] P. Lutze, R. Gani, J.M. Woodley, Process intensification: a perspective on process synthesis, *Chem. Eng. Process.: Process Intensif.* 49 (6) (2010) 547–558.
- [95] J.D. Perkins, S.P.K. Walsh, Optimization as a tool for design/control integration, *Comput. Chem. Eng.* 20 (4) (1996) 315–323.
- [96] J. van Schijndel, E.N. Pistikopoulos, Towards the integration of process design, process control and process operability: current status and future trends, in: M.F. Malone, J.A. Triandam, B. Carnahan (Eds.), Proceedings of the 5th International Conference on Foundations of Computer-Aided Process Design, Elsevier, New York, 2000, pp. 99–112.
- [97] V. Sakizlis, J.D. Perkins, E.N. Pistikopoulos, Recent advances in optimization-based simultaneous process and control design, *Comput. Chem. Eng.* 28 (10) (2004) 2069–2086.
- [98] D.R. Lewin, Interaction of design and control, in: Proceedings of the 7th IEEE Mediterranean Conference on Control and Automation, Haifa, Israel, 1999.
- [99] V. Bansal, J.D. Perkins, E.N. Pistikopoulos, R. Ross, J.M.G. van Schijndel, Simultaneous design and control optimisation under uncertainty, *Comput. Chem. Eng.* 24 (2–7) (2000) 261–266.
- [100] C.P. Almeida-Rivera, J. Grievink, Process design approach for reactive distillation based on economics, exergy, and responsiveness optimization, *Ind. Eng. Chem. Res.* 47 (1) (2008) 51–65.
- [101] M. Miranda, J.M. Reneaume, X. Meyer, M. Meyer, F. Szigeti, Integrating process design and control: an application of optimal control to chemical processes, *Chem. Eng. Process.: Process Intensif.* 47 (2008) 2004–2018.
- [102] M.C. Georgiadis, M. Schenk, E.N. Pistikopoulos, R. Gani, The interactions of design, control and operability in reactive distillation systems, *Comput. Chem. Eng.* 26 (4–5) (2002) 735–746.
- [103] H.A. Preisig, Constructing and maintaining proper process models, *Comput. Chem. Eng.* 34 (9) (2010) 1543–1555.
- [104] M.R. Westerweele, J. Laurens, Mobatec modeller—a flexible and transparent tool for building dynamic process models, *Comput. Aided Chem. Eng.* 25 (2008) 1045–1050.
- [105] L.T. Biegler, I.E. Grossmann, Retrospective on optimization, *Comput. Chem. Eng.* 28 (8) (2004) 1169–1192.
- [106] L.T. Biegler, *Dynamic Optimization Methods with Embedded DAE Solvers (Chapter 9) in Nonlinear Programming: Concepts, Algorithms, and Applications to Chemical Processes*, Society for Industrial and Applied Mathematics (SIAM), Philadelphia, PA, 2010.