An Overview of Process Intensification Methods

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Abstract

Process Intensification promises novel solutions to current challenges in the chemical process industry, leading to a rapid growth in interest. There are different approaches to synthesize an intensified process, most of which are based on methods from Process Synthesis and Process Optimization. In this paper, we review those methods and provide an overview of their application in Process Intensification. We also review the evolution of phenomena-based representations, a central theme in the synthesis for PI. Finally, we summarize and compare the approaches that have been suggested for retrofit using PI and criteria to evaluate PI options.

Highlights

Comparison of Process Synthesis (PS), Process Optimization (PO) and Process Intensification (PI)
Review and evaluation of methods to perform PS, PO, and PI
Literature survey on Methodologies for Systematic PI Incorporation
Evaluation Methods for PI
Mathematical Optimization shows great promise towards the development of PI

1. Introduction

Facing numerous challenges such as minimizing energy and waste generation or improving economic as well as environmental metrics, the chemical industry constantly seeks new solutions to fulfill ever-evolving demands. Process Intensification (PI) promises long-term solutions to these challenges, thus research interest has recently rapidly grown, with more than 12 books published since 2003 [1-12]. Many excellent reviews have been published summarizing the last developments [13-15]. Major examples of Process Intensification include reactive distillation, rotating packed beds and microreactors. Reactive distillation combines reaction and separation by filling a distillation column with a catalyst [16] for a simultaneous removal of vapor products from a boiling, reacting mixture [17]. Thus, the equilibrium is pulled to higher conversions [18], thereby enhancing overall reaction rates as well as selectivity [17] while reducing energy needs by 80% and investment cost by 20% [19]. Rotating packed beds are excellent devices to overcome mass-transfer limited processes, utilizing centrifugal acceleration [20] to create uniform dispersion and high turbulence [4]. Consequently, the volumetric mass transfer coefficient can be increased by 1-2 orders, reducing the necessitated volume by 91% compared to a conventional packed bed [20]. Microreactors are chemical reactors of extremely small dimensions. Through their characteristic low reaction-volume to surface area ratios, they allow very high heat transfer rates [21] as well as an excellent control of reaction temperature [22] alongside with safer processes that include poisonous or explosive reactants. In 2009, DSM substituted a conventional reactor with a microreactor, scaling its content down from $10m^3$ to $0.003m^3$, improving the selectivity as well as the material yield by 20% while still achieving the production goal [23, 24].

Similar to PI and for related reasons, Process Optimization (PO) and Process Synthesis (PS) have likewise experienced a great rise in interest among the research community ever since the turn of the millennium (see Figure 1). PO benefits from significant advances in the speed and robustness of NLP and MINLP algorithms. Moreover, powerful optimization modeling environments enable the formulation and solution of large-scale optimization applications [25]. Excellent reviews can be found in [26, 27]. PS has notably witnessed great advances in synthesis tools and techniques, mostly for heat exchangers and separation networks. Recent reviews of this field of study have been provided by Cremaschi et al. (2015) [27] as well as Chen and Grossmann (2017) [28].

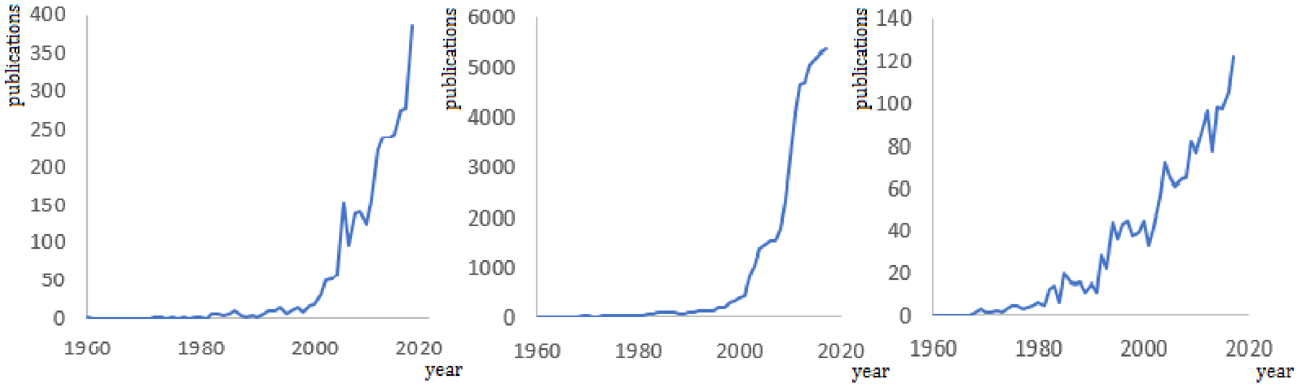

Figure 1: Scopus: Contributions on PI, PO and PS (from left to right) between 1960 and 2017

Despite extensive research in PI, it still lacks a single canonical definition. PI was traditionally understood as process development leading to reduction in equipment size [29]. Recently, PI has been extended to include business, process, and environmental aspects [29] and is regarded as a holistic approach to improve a process. For this work, we will adopt the definition from Cross and Ramshaw (1986), according to which PI is a methodology for making remarkable reductions in equipment size, energy consumption, or waste generation while achieving a given production goal [30].

Even though PI, PO and PS are not the same field of study (Figure 2), their development and advances correlate, as they share commonalities in methodology.

	PO	**PS**	**PI**
Aim	Performance improvement of existing concepts	Assembly of process network to optimize either economic, environmental, and/or social objectives	Development of new concepts of process steps and equipment
Focus	Model, numerical method	Superstructures	Experiment, phenomenon, interface
Interdisciplinarity	Interface with applied math and mathematical programming)	Mathematical programming, informatics, chemistry, physics	Strong (chemistry and catalysis, applied physics, mechanical engineering, materials science, electronics, etc)

Figure 2: Basic features of Process Optimization (PO), Process synthesis (PS), Process Intensification (PI) (adopted from Keil 2018 [31]).

1. 1 Methods for Process Optimization, Process Synthesis and Process Intensification

Process Optimization

relies on mathematical optimization, employing an objective function to minimize or maximize. This objective function is constrained by the feasible region of the system performance, defined by process specifications and physical relations with respect to the system variables. Optimization-based methods utilize mathematical programming for overall flowsheet synthesis [32].

Methods to perform **process synthesis** as well as **process intensification** have been classified into heuristic (and knowledge-based), mathematical optimization, and hybrid methods.

1.1.1 Heuristic

The heuristic approach is based on rules gained by experience, as well as process insights at the unit operations scale [33] and verified through simulation or experimentation. The three main categories of heuristics methods are data models, data mining models, and application models. Heuristic methods are helpful in recommending process improvements for existing process (retrofit) [32] but limited in their generality for diverse processes.

1.1.2 Mathematical optimization:

Optimization-based methods necessitate the generation of a superstructure, a representation comprised of all plausible flowsheet alternatives. The superstructure is translated into an MINLP optimization problem with an objective function typically pertaining to process economics and subject to constraints for the operating conditions. By simultaneously optimizing the flowsheet structure and operating conditions, an improved design can be generated than with a sequential approach. The limitations, however, are in the formulation of a tractable MINLP, sometimes necessitating the introduction of simplifying assumptions that reduce the design space or modeling fidelity. Similarly, the superstructure must be generated to embed the optimal pathway or it will not be found [32-35].

1.1.3 Hybrid

Hybrid methods attempt to combine the advantages of both heuristic as well as mathematical optimization methods [33]: they keep the simple structure of the heuristic approach whilst replacing the fixed rules with thermodynamic insights [36], therefore narrowing the search space by removing physically impossible or improbable solutions. While this screening process may prematurely remove an optimal solution, the result is often a smaller MINLP or NLP problem, significantly reducing the computing effort required for the design problem.

	Heuristic [52-55]	**Mathematical Programming [26 ,55- 57]**	**Hybrid [39, 55, 58- 61]**
Methodology	Knowledge-based methods from sets of heuristic rules	Optimization problem using a generated superstructure based on engineering judgment	Structure of heuristic method but replacing fixed rules with thermodynamic insights
Scale	Unit Operations Scale Task Scale	Unit Operations Scale Task Scale	Unit Operations Scale Task Scale Phenomena Scale
Advantages	- Simple, fast - Gives recommendations for improvement of existing process or new process similar to studied one	- Simultaneous optimization provides better results than sequential	- Simple structure (heuristic approach) - Narrows search space by thermodynamic insights - Less complex MINLP/NLP problem
Limitations	- Lack of generality - Requires extensive expert knowledge	- MINLP simplification may preclude new solutions - Optimal pathway must already be implied in original superstructure	- Heuristic screen may eliminate non-intuitive optimal solutions
Intensification of one Process Section	Bessling et al., 1997 [37] Kiss et al., 2007 [38]	Amte, 2011 [41] Caballero and Grossmann, 2004 [42] Urselmann et al., 2011 [43] Ramapriya et al., 2014 [44] Anantasarn 2017 [45] Chen and Grossmann, 2017 [20]	Freund and Sundmacher, 2008 [48] Peschel et al., 2012 [49] Seifert et al., 2012 [50]
Intensification of Entire Process	Siirola, 1996 [39] Portha et al, 2014 [40]	Papalexandri and Pistikopoulos, 1996 [46] Demirel et al, 2017 [47]	Lutze et al., 2013 [36] Babi et al., 2014 [51] Tula et al., 2017 [33]

Figure 3: Overview of Process Synthesis-Intensification Methods and their features

2. A holistic and systematic framework for intensification

Most of these methods focus on intensification methods at the unit operation scale that improve a certain set of tasks and/or phenomena, leaving the interactions between this particular unit operation and the rest of the process out of focus [34]. In this way, the impact of local intensification of a single unit can be very limited, resulting in weak improvement of the whole process [40]. This holistic view on process intensification was discussed by Freund and Sundmacher (2008) [48] as well as by Ponce-Ortega (2012) [62]. As an alternative, plant intensification considers the interactions among all units within the process. Their mathematical model implies a superstructure approach based on previously developed superstructures [63-72], utilizing the concept of path equations [73]. Resulting in a general mathematical model, researchers can accordingly intensify for either one existing or added unit or an entire plant. This mathematical model, however, was criticized as lacking realism by Portha et al. [40]. They differentiated between local and global process intensification with the former applying to the classical approach of PI, intensifying a single unit. Global PI simultaneously improves several units and also takes different drivers such as economic, safety, eco-efficiency and sustainability into account, therefore combining PS, PI and PO. Since mathematical process models enable overall flowsheet synthesis, their use is essential for a systematic and efficient process design [32]. The necessary multi-objective optimization techniques were studied by [74-76]. These mathematical optimization-based approaches are advantageous for studying an entire process. They enable researchers to systematically intensify a large scale process and quantify the achieved process improvement.

2.2 Bottom-Up Approaches (phenomena-based)

The latest trend in holistic systematic process intensification involve “bottom-up” approaches. In these approaches, researchers depart from conventional unit operations, regarded as the highest level of aggregation [77], and from pre-specified process alternatives that restrict the design search space. These so-called “ready-solutions” [48] limit the researchers’ creativity to find novel intensified tasks or equipment and narrow the design space [77, 78]. Instead, they propose to decompose the entire process into a set of processing tasks, physiochemical phenomena and functions [46]. Starting at a lower level of aggregation, chemical processes are decomposed into a combination of several phenomena such as cooling, heating, mixing, reaction, dividing etc. which are thereafter re-grouped back into unit operations, promoting the exploitation of PI options [55]. In addition to that, the phenomenological descriptions can be directly translated into mathematical equations [79], enabling mathematical programming-based optimization to simultaneously identify the best design, synthesis and intensification routes [47], hence it can outperform task- or equipment based approaches [32].Considering the trade-offs among different parts of the process, optimization-based methods provide a more holistic process synthesis and intensification as it can handle the respective variables simultaneously [32]. Since complexity of the resulting optimization problem presents a significant challenge [77], a trade-off is required between the number of combinations included in the superstructure, the tractability of the (MINLP) problem, and the uniqueness of the solution [49].
Based on these ideas, many methodologies have been proposed. We summarize these in Figure 4.

Reference	Method	Annotation
Papalexandri and Pistikopoulos, 1996 [46]	Phenomena-based generalized modular representation framework (GMF) that optimizes mass and heat transfer performances based on Gibbs free energy, eventual solution of an optimization-based superstructure problem with MINLP formulation	Framework has been applied for -combined separation/reaction systems -azeotropic separation systems multicomponent separation systems -heat-integrated distillation systems Criticism: model simplification to handle combinatorial complexity of problem might result in infeasible solutions. [77]
Arizmendi-Sánchez and Sharratt, 2008 [80]	Hybrid qualitative (knowledge based)-quantitative (causal graphs) approach: Physicochemical phenomena arranged into equipment-independent functional, structural, behavioral modules	Criticism: does not describe the modeling of multiple physical scales, thus not defined where process intensification measures are included
Rong et al., 2008 [81]	Phenomena-based approach to use more physical and chemical insights Limiting steps of process identified and replaced by better suited equipment	Criticism: limited to retrofit problems
Freund and Sundmacher, 2008 [48] **Peschel et al., 2010 [82]** **Peschel et al., 2011 [83]**	Equipment-independent process flowsheet composed of task-based “functional modules” that can be further decomposed to linear combination of elementary process functions and flux vectors Systematic identification and investigation of suitable measures for PI	Application-focus on reactor design and synthesis
Lutze et al., 2013 [36]	Database of phenomena that are combined to form simultaneous phenomena building blocks (9 major classes) from which phenomena-based flowsheet variants are generated by series of screening steps and connection rules (3-stage-approach)	Framework applied to produce isopropyl acetate -extended by Babi [51] for membrane-based processes with sustainability considerations
Babi et al., 2014 [84]	Based on Lutze et al.‘s [36] framework: translation of phenomena-based flowsheets to innovative designs (superstructure-based process synthesis) plus addition of sustainability analysis	Continued work by Kuhlmann and Skiborowski [85]: proposition of state-space superstructure for systematic generation of flowsheet variants Complementary computer-aided work-flow proposed by Anantasarn et al., 2017 [45]
Demirel et al., 2017 [47]	Building blocks mimicking fundamental phenomena where assembly of different types of blocks result in an intensified unit no a priori postulation of potential process configurations Overall design problem as single MINLP problem 2-Dgrid representation	Automatically generates intensified flowsheets Extended to include capital cost considerations [86] and process integration [87] Tailored computer-aided framework “SPICE” [88]

Figure 4: Overview of the evolvement of phenomena-based approaches

2.3 Retrofit

The methodologies presented in Figure 4 mainly relate to green field constructions. However, we also need to consider the option of incorporating Process Intensification in an existing process through process retrofitting (PR).

The industrial motivations to retrofit a process are numerous: increasing productivity, capacity and safety whilst decreasing energy usage, waste and operational costs [79]. Grossmann et al. (1987) [89] estimates that 70-80% of all process design projects deal with retrofit. Their definition of process retrofit shares many objectives with Process Intensification and Process Systems Engineering, such as decreased energy consumption and improved quality, conversion and safety, suggesting a synergistic relationship.

To our best knowledge, only two works address retrofit design using PI technology. The first approach is presented by Niu et al. (2016) [90] as a four step heuristic methodology including a base case analysis, the generation of an improved solution without capital investment, the generation of integrated solutions, optimization and eventually the comparison of solutions. This contribution focuses mainly on the integration of units, in particular reactive as well as hybrid separations.

The second, Barecka et al. (2017) [91], presents a methodology consisting of process analysis, bottleneck identification and selection of the most promising PI option from a database to quickly quantify the improvement.

While these two methodologies pursue the same aim of supplying the research community with a useful tool for retrofit incorporating PI strategies, they differ in their respective approaches, advantages, and limitations. Niu et al. (2016) relies on heuristic screening and intensification by integration. A series of sequential analyses generate intensified process alternatives, giving practitioners a simple avenue to explore PI improvements. However, the quantitative assessment of alternatives necessitates tedious simulations, hence, a very large effort for the systematic evaluation of various process options.

Barecka et al. (2017) base their method on a PI database with more than 150 intensified technologies. By utilizing metrics for benchmarking different retrofit strategies, merely the most promising options are subsequently considered which lessens the simulation effort drastically compared to the procedure in Niu et al. (2016). However, their limitation lies in the in-depth insight into the physiochemical phenomena causing the bottlenecks. Those necessitate detailed data from literature which might not always be at hand.

These heuristic (Niu et al., 2016) [90] and hybrid (Barecka et al., 2017) [91] methodologies enable us to implement process intensification into process retrofit. While these approaches offer compelling options to practitioners, there is scope for compromise alternatives. A mathematical optimization-based strategy would complement existing approaches and open up new opportunities for this field of study.

3. Criteria to evaluate PI

Despite potential advantages, new PI technology faces many barriers to implementation, such as risk due to lack of precedent, concerns about safety and control, and considerable time-to-market due to exhaustive data acquisition and simulation [79, 92].We need tools to evaluate and possibly quantify the overall performance of a novel intensified process. The difficulty in quantifying the improvement of independent factors that do not directly relate to cost (e.g. safety, non-technical factors) is discussed by many researchers [31, 79, 93, 94]. A popular method is the application of intensification metrics. The most widespread PI metrics for evaluation are classified into four indicators: economic, environmental, safety, and intrinsic intensified [79]. Those indicators are measured, weighted according to their social and environmental impact [79, 95] and eventually compared [31]. This method has been applied with slight variations by numerous authors [34, 74, 96, 97].The European Roadmap benchmarked different PI technologies through their performance with respect to transfer phenomena [98]. Inspired by green chemistry evaluation tools [99, 100] and other contributions [101], Rivas et al. (2018) [102] propose the “intensification factor”, an arithmetic method to evaluate PI options. It is composed of modular interchangeable evaluation criteria and can combine qualitative as well as quantitative factors such as economical, technical, scientific aspects. The strongest feature is its simplicity that allows users to quantify the factor with minimal information during an early design stage and can be understood by outsider and non-experts. Finally, mathematical optimization does not only act as a tool for overall flowsheet synthesis to generate intensified processes as discussed earlier. It also provides a quantitative evaluation through its objective function, thereby facilitating the final decision with regard to the process alternatives [36, 47, 79, 84].Therefore, it seems to be one of the most promising approaches towards the implementation of PI.

4. Conclusion

Process Intensification attracts broad interest for its promise in solving current challenges in the chemical process industry. To bring its benefits into practice, engineers need to synthesize new, intensified processes. Several different approaches exist to synthesize an intensified process, many of which have roots or parallels in Process Synthesis and Process Optimization. Among them, Mathematical Programming is a powerful tool, allowing practitioners to postulate new PI solutions at the level of fundamental phenomena and then to systematically evaluate their potential. However, there are still many open challenges, such as the complexity in finding a global solution to the MINLP synthesis problem, and the generation of an appropriate initial superstructure.

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