

Juan Gabriel Segovia-Hernández
Adrián Bonilla-Petriciolet *Editors*

Process Intensification in Chemical Engineering

Design Optimization and Control

 Springer

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ISBN 978-3-319-28390-6 ISBN 978-3-319-28392-0 (eBook)
DOI 10.1007/978-3-319-28392-0

Library of Congress Control Number: 2016931878

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Preface

Today, we are witnessing important new developments that go beyond traditional chemical engineering. Engineers and industrial researchers are working on novel equipment and techniques that potentially could transform our concept of chemical plants and lead to compact, safe, energy-efficient, and environment-friendly sustainable processes. These developments share a common focus on “process intensification,” an approach that has been around for quite some time but has truly emerged only in the past few years as a special and interesting discipline of chemical engineering.

Process intensification can be defined as: “Any engineering development that leads to a substantially smaller, cleaner, safer, and more energy-efficient technology.” The application of this concept in process system engineering is most often characterized by a significant reduction in plant volume, production costs, waste generation, and also getting improvements, even in orders of magnitude, on process performance and efficiency including the reduction of environmental pollution problems. In recent years, process intensification has attracted considerable academic interest as a potential means for process improvement and to meet the increasing demands for a sustainable production. A variety of intensified operations developed in academia and industry creates a large number of options to potentially improve the process. However, the task for identifying the set of feasible solutions for process intensification in which the optimal can be found may take considerable resources. Hence, a synthesis tool to systematically achieve the process intensification would potentially assist in the generation and evaluation of process options.

Currently, several process design tools with a clear focus on specific process intensification tasks exist. Therefore, this book covers current topics for the design, optimization, and control in the context of process intensification. This book was motivated by the desire we and others have had to show the evolution and advances in this area. Chapters of this book cover a variety of concepts and aspects, involving a variety of processes as case of study and examples, related to process intensification in chemical engineering.

We are deeply indebted to the authors who have contributed to this book. Also, we acknowledge the anonymous reviewers for their time and effort in assisting us with the edition of this book.

Guanajuato, Mexico
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Juan Gabriel Segovia-Hernández
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Chapter 1

Introduction

Juan Gabriel Segovia-Hernández and Adrián Bonilla-Petriciolet

Abstract This chapter provides an overview of process intensification in chemical engineering and summarizes the content of this book.

The chemical, pharmaceutical, and bio-based industries produce products that are essential for modern society. Nevertheless, these industries face considerable challenges because of the need to develop sustainable production methods for the future.

Process intensification (PI) targets dramatic improvements in manufacturing and processing by rethinking existing operation schemes into ones that are both more precise and efficient than existing operations. PI frequently involves combining separate unit operations such as reaction and separation into a single piece of equipment resulting in a more efficient, cleaner, and economical manufacturing process. At the molecular level, PI technologies significantly enhance mixing, which improves mass and heat transfer, reaction kinetics, yields, and specificity. These improvements translate into reductions in equipment numbers, facility footprint, and process complexity, and, thereby, minimize cost and risk in chemical manufacturing facilities.

In the frame of globalization and sustainability, the future of chemical engineering can be summarized in four main objectives:

1. Increase productivity and selectivity through intensification of intelligent operations and a multiscale approach to processes control (e.g., nano- or microtailoring of catalyst materials).
2. Design novel equipment based on scientific principles and new production methods: process intensification in using multifunctional reactors, microengineering, and microtechnology.

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3. Design and engineering using the “triplet molecular Processes-Product-Process Engineering)” approach to manufacture end-use properties.
4. Implement multiscale application of computational chemical engineering modelling and simulation to real-life situations from the molecular scale to the production scale [1].

At the core of PI is the optimization of process performance by focusing on molecular level kinetics, thermodynamics, and heat and mass transfer. Gerven and Stankiewicz (2009) provide four guiding principles for PI [2]:

1. Maximize effectiveness of intramolecular and intermolecular events (e.g., dynamically changing conditions to attain kinetic regimes with higher conversion and selectivity).
2. Provide all molecules the same process experience (e.g., plug flow reactor with uniform, gradient-less heating).
3. Optimize driving forces at all scales and maximize the specific surface areas to which they apply (e.g., increase transfer surface area through microchannel designs).
4. Maximize synergistic effects from partial processes (e.g., affecting reaction equilibrium by removing products where and when they are formed).

PI designs that achieve all or some of these molecular-level optimal conditions are likely to be transformative. Reactors that enable precise control of the reactor environment could dramatically increase yields, conversions, and selectivity, which in turn would reduce material, energy, and carbon intensities, minimize purification needs, and reduce waste disposal burdens. Additionally, PI technologies could enable the manufacture of products that otherwise could not be safely or successfully made.

In general, process intensification consists of the development of novel apparatuses and techniques that, compared to those commonly used today, are expected to bring dramatic improvements in manufacturing and processing, substantially decreasing equipment-size/production-capacity ratio, energy consumption, or waste production, and ultimately resulting in cheaper, sustainable technologies. Or, to put this in a shorter form: any chemical engineering development that leads to a substantially smaller, cleaner, and more energy efficient technology is process intensification [3].

The whole PI field generally can be divided into two areas:

- Process-intensifying equipment, such as novel reactors, and intensive mixing, heat-transfer and mass-transfer devices.
- Process-intensifying methods, such as new or hybrid separations, integration of reaction and separation, heat exchange, or phase transition (in the so-called multifunctional reactors), techniques using alternative energy sources (light, ultrasound, etc.), and new process-control methods (like intentional unsteady-state operation). Obviously, there can be some overlap. New methods may

require novel types of equipment to be developed and vice versa, while novel apparatuses already developed sometimes make use of new, unconventional processing methods.

Commercial applications of PI date back to the 1970s. Static mixers, which are ubiquitous today, were early PI inventions. Other early PI technologies deployed reactive distillation, including Eastman Chemical Company's tower reactor, which integrated five processing steps in the production of methyl acetate from methanol, achieving an 80 % reduction in energy and a large reduction in capital costs. In the chemical industry, reactive distillation, divided wall column distillation, and reverse flow reactors have been commercialized each with more than 100 installations. Drivers for PI innovation include the potential for reduction in feedstock cost, capital expenditure, energy, and safety issues. Barriers to deployment include risk of failure, scale-up unknowns, unreliability of equipment, and uncertain safety, health, and environmental impacts [4].

In recent years, PI has attracted considerable academic interest as a potential means for process improvement to meet the increasing demands for sustainable production. Process intensification is gaining much attention as one of the key objectives in designing new plants and retrofitting existing units. Several drivers have contributed to this increasing attention. For instance, enhanced process safety and homeland security are tied to process intensification; as the inventory and flows of hazardous substances are lowered, the process risk is typically reduced. As many processes, particularly those in the chemicals, nuclear and oil industries, involve the production, handling, and use of hazardous substances, process intensification is one way in which the inventory of such substances, and the consequences of a process failure, may be significantly reduced. PI, therefore, has the potential to be a significant factor in the implementation of inherent safety. Additionally, conservation of natural resources (including better utilization of mass and energy), biotechnological applications, and new control methods may be linked to Process Intensification. Also, PI is a valuable approach in developing economical processes with a minimal global footprint which will require new infrastructure to be designed and built [5]. PI is in development and, as is often the case in the emergence of new areas, there is an ongoing discussion about its definition. Consensus has been reached that PI involves creative innovation and stimulates the engineering community to strive to real breakthroughs, aiming at plants that are more compact, cost-effective, and safer. An integrated (or holistic) approach is essential, resulting in a symbiosis with well-defined disciplines, in particular Process Systems Engineering [6].

This book gathers research from across the globe in the study of design, control, and optimization in process intensification. Topics discussed include process separation, bioprocess, heat and mass exchanger networks, industrial applications, etc.

Chapter 2 gives an overview of the fundamentals of process intensification from a process systems engineering point of view. The concept of process intensification, including process integration, is explained together with the drivers for applying process intensification, which can be achieved at different scales of size, that is, the unit operation scale, the task scale, and the phenomena scale.

In Chapter 3, a systematic methodology to generate different classes of distillation configurations is presented. The generation methodology is able to consider thermally coupled, thermodynamically equivalent structures and intensified alternatives with a less number of columns compared to the corresponding simple column sequences. The methodology described has the advantage to produce a complete set of alternatives, avoiding the trial-and-error procedure with random configurations picked up from the literature. Finally, the methodology described has the benefit to keep a clear connection between the simple column sequences and all the alternatives predicted. This aspect helps the designer in the definition of columns' configuration parameters.

Chapter 4 presents the use of process integration as a useful tool for intensifying processes. Particularly, mass and heat integration through the synthesis of mass and heat exchanger networks represent powerful tools that can be used for reducing the need of external agents such as fresh water and hot and cold utilities. Two optimization formulations are presented for mass and heat integration and the application to two case studies show significant savings of external utilities.

Heat integration between vapor and liquid streams has been widely used in chemical and petrochemical plants for conventional distillation processes as an alternative to reduce the energy consumption. However, with the advances that have been proposed in intensified distillation processes in the last couple of decades, heat-integrated alternatives that are more attractive than the typical condenser–reboiler heat integration have also been proposed. Therefore, intensified distillation processes also need a new approach methodology to implement optimal locations and heat load in heat-integrated distillation. Chapter 5 aims to cover the fundamentals, simulation and optimization approaches for heat-integrated intensified distillation processes for nonreactive and reactive systems.

Reactive distillation stands out as a successful example of a process intensification technology for enhanced chemical manufacture. After almost a century of development, it has achieved a high degree of maturity in terms of design capabilities, the availability of commercial suppliers of hardware and software, and a large variety of processes effectively implemented at the industrial scale. Based upon an extensive review of the classical and recent literature on reactive distillation.

Chapter 6 briefly describes the context in which the technology was developed, its current status, and the expected areas for progress in the near future. In addition, the intensification principles behind the operation, its fundamentals, constraints, design methodologies, the optimization approaches, and the control strategies are discussed with fair detail. Finally, a case study on the ethyl acetate production via esterification of acetic acid with ethanol by reactive distillation is presented. In this example, a complete process synthesis procedure is described, from the conceptual design all the way to the process optimization.

In Chapter 7, bioprocess intensification is explored mainly from the perspective of transforming biomass into chemicals as an integrated solution for bioprocessing. Then, bioprocess intensification is addressed within the biorefinery context.

Chapter 8 presents process intensification technologies used in industrial applications, for increasing the eco-efficiency of the chemical equipment with the benefit of lower capital costs, substantial energy saving, reduced footprint, and safety by design. The key topics cover compact heat exchangers, static mixers, green chemical reactors (e.g., micro-reactors), high-gravity (HiGee) technology, cyclic distillation, dividing-wall column, and reactive distillation.

Chapter 9 describes and discusses stochastic optimization methods for solving problems involved in process intensification, given an emphasis in multi-objective optimization due to its increasing importance in the chemical engineering community. A brief description of the multi-objective optimization strategies such as genetic algorithms, simulated annealing, tabu search, differential evolution, ant colony and particle swarm optimization is provided, including several applications of evolutionary optimization methods in the intensification of separation processes.

Process intensification is a branch of Chemical Engineering which has taken importance in the last decades because through its application it is possible to obtain alternative processes with smaller/multitask equipment and reduced energy requirements. Such reductions may have a positive benefit to the environment, since smaller equipment implies less use of material for its construction; while reductions on energy requirements implies lowering direct emissions of greenhouse gases to the atmosphere. In Chapter 10, examples of intensification alternatives recently proposed for enhancement of processes is presented. In particular, process intensification in the production of liquid biofuels is analyzed. The application of such tool in the production of biofuels, which are expected to reduce environmental impact when compared to the use of fossil fuel, has both energy savings and further reductions in terms of pollutant emissions.

The arts of design, optimize, and control of chemical processes should be considered simultaneously. Nevertheless, in the case of process intensification the most common situation is that design is performed as first stage (following mass/energy integration guidelines), secondly processes are optimized (costs, profit, environmental impact), and finally a control scheme is adopted. Additionally, it is necessary to consider that intensification generates new process dynamics (different responses and characteristic times) and reduces, notoriously, the number of manipulate variables available for control. Hence, the original difficult tasks of partial control and stability of both, process and control, becomes more complicated. Chapter 11 is devoted to the analysis of the problems mentioned above, which are inherent to any chemical process although more evident during process intensification. Some special features of the control are identified and some suggestions are given to enface problems that arise after the intensification of some separation and reaction/separation examples.

In this book, we have highlighted a variety of topics that should play a significant role in the intensification of chemical processes. This has not been a comprehensive cataloging, as new developments are regularly emerging from researchers worldwide. The usage of a knowledge base has been presented to provide data for various PI applications, chemical systems, conditions, etc.

Process intensification, although yet to fully emerge as an established technology in the process industries, offers significant opportunities in sectors ranging from chemicals to food. The possibility of increased level of production and the growing use of biological renewable feedstocks opens up new challenges and opportunities for those active in integration as well as intensification.

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Chapter 2

Fundamentals of Process Intensification: A Process Systems Engineering View

Deenesh K. Babi, Mauricio Sales Cruz, and Rafiqul Gani

Abstract This chapter gives an overview of the fundamentals of process intensification from a process systems engineering point of view. The concept of process intensification, including process integration, is explained together with the drivers for applying process intensification, which can be achieved at different scales of size, that is, the unit operation scale, the task scale, and the phenomena scale. The roles of process intensification with respect to process improvements and the generation of more sustainable process designs are discussed and questions related to when to apply process intensification and how to apply process intensification are answered through illustrative examples. The main issues and needs for generation of more sustainable process alternatives through process intensification are discussed in terms of the need for a systematic computer-aided framework and the methods and tools that should be employed through it. The process for the production of methyl-acetate is used as an example to highlight the generation of more sustainable process alternatives through this framework. Perspectives, conclusions, and future work are proposed in order to further develop the field of process intensification using a systems approach.

2.1 Introduction

The objective of process synthesis is the selection of the best (optimal) process flowsheet from among numerous alternatives for converting specified raw materials into specific desired products, subject to predefined performance criteria [1]. More

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J.G. Segovia-Hernández, A. Bonilla-Petriciolet (eds.), *Process Intensification in Chemical Engineering*, DOI 10.1007/978-3-319-28392-0_2

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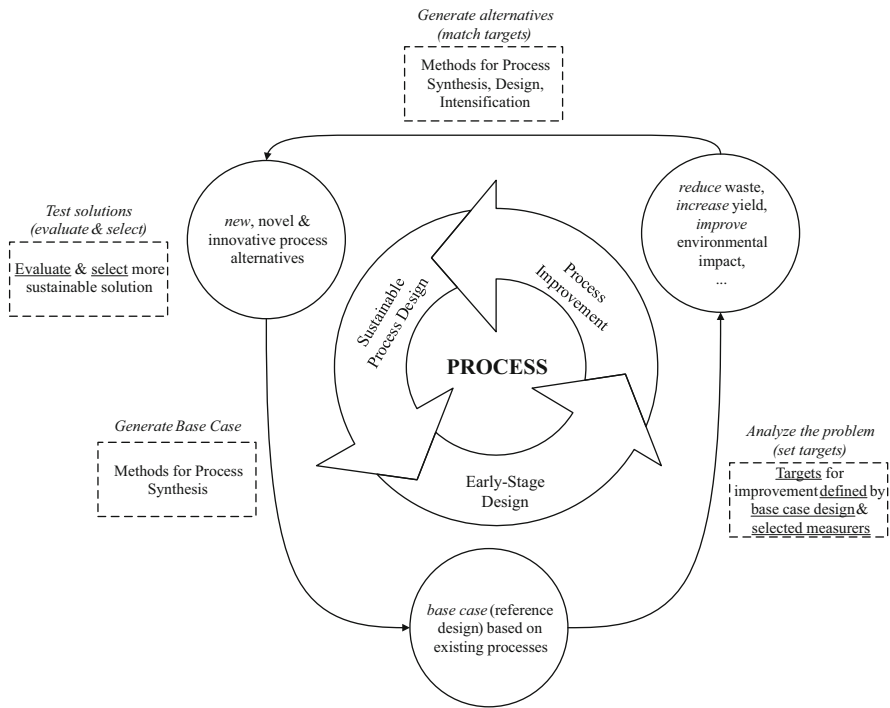


Fig. 2.1 Role of process intensification within sustainable process design

sustainable design is defined as the design of process flowsheets that correspond to lower values of a set of targeted performance criteria based on economical, operational, and environmental factors [2]. As shown in Fig. 2.1, the sustainable design is obtained through improvements with reference to the early stage design (also called reference design or base-case design). The role of process synthesis in finding the early stage design and/or the more sustainable design is to generate the feasible process alternatives. The generation of alternatives can be done in various ways, for example, trial and error, rule-based heuristics, process integration (mass and energy), process optimization, process intensification, and many more. In this chapter, process intensification, which also includes process integration, is presented. Through the improvements as shown in the middle layer of Fig. 2.1, the following improvements related to the physical system can be obtained through process intensification [3]:

- Catalysts—the screening and selection of novel catalysts for reactive systems
- Solvents—the generation, screening, and selection of environmentally friendly solvents for separation processes
- Materials—the design of new materials for the design of novel, innovative unit operations that combine reaction, separation, or reaction–separation systems

- Energy—the use of process integration concepts to generate more sustainable process alternatives with respect to reduced waste and environmental impact

Process intensification has been receiving increased attention and importance because of its potential to obtain innovative and more sustainable process design alternatives. But what is process intensification? Many definitions have been proposed for process intensification and a few are highlighted here. Stankiewicz and Moulijn [4] defined process intensification as the development of novel and sustainable equipment that compared to the existing state-of-the-art, produces dramatic process improvements related to equipment sizes, waste production, and other factors. Reay et al. [5] defined process intensification as process development that involves reduction in equipment (unit operation) sizes that lead to improvements in reaction kinetics, better energy efficiency, reduction in capital cost, and improvement in process safety. Ponce-Ortega et al. [6] defined process intensification as an activity characterized by five principles—reduced size of equipment, increased throughput of process, reduced equipment holdup or inventory, reduced usage of utilities and raw materials, and, increase efficiency of process equipment. The above definitions provide insights into the different scales at which process intensification can be employed.

With the exception of process integration, there are, however, not many published methods to determine the intensified solutions for processes, even though successful intensified solutions have been reported by many. Examples of process intensification methods have been proposed at unit operation scale by Bessling et al. [7], at task scale by Agreda et al. [8] and at phenomena/molecular scale by Freund and Sundmacher [9]. These methods however, mainly focus on the design of new/novel unit operations that enhances/integrates a particular set of tasks and/or phenomena within the process, but does not consider the interaction of the rest of the unit operations that constitute the final (total) process design. They are also specific to the characteristic of the unit operations studied.

With a systematic and generic method to find process intensification solutions, Lutze et al. [10] recently proposed a definition for process intensification that covers all three scales and consider the overall process. According to Lutze et al. [10], process intensification is defined as the targeted improvement of a process at the unit operations scale, the task scale, and/or the phenomena scale. With overall improvement as the objective, at the unit operations scale the individual equipment that constitute the final process design of any chemical or biochemical process is identified and/or designed; at the task scale, the functions (tasks) performed by each unit operation are identified and analyzed; and, at the phenomena scale, the phenomena that satisfies the tasks to be performed are identified and analyzed. In this way, links between the scales are established and allows the search in various scales to find the design of new, innovative, and sustainable processes. This method can be applied to the design of new processes as well as, the retrofitting of existing processes, in order to make them more sustainable. This concept of scales is illustrated through Fig. 2.2 where at the lower scales, a small set of tasks and phenomena are employed to represent various

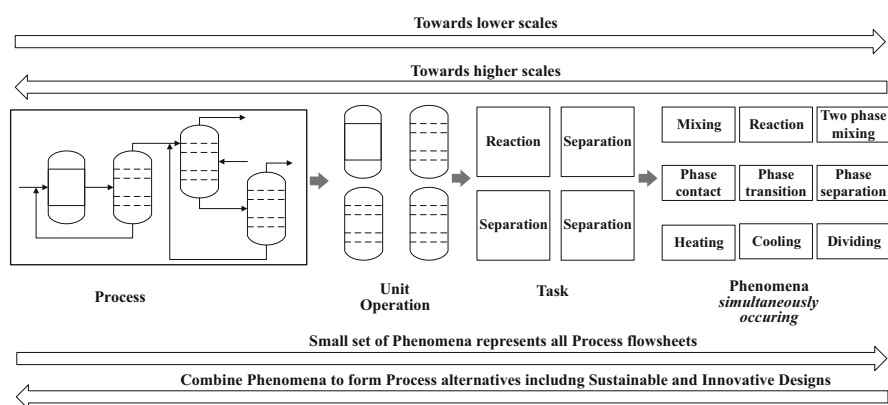


Fig. 2.2 Multiscale method for process intensification

process flowsheet alternatives. Moving from the higher to the lower scale increases the possibility for process innovation because of the following reasons:

- New alternatives that match the functions of unit operations can be generated through the combination of tasks, leading to new flowsheet alternatives.
- New ways to perform tasks can be generated by combining different phenomena, leading to the generation of novel and (more sustainable) flowsheet alternatives.

Therefore, if processes are designed at scales lower than the unit operation scale, there is an increased chance of finding a more sustainable overall process design. This is highlighted by the process intensification solutions reported by Agreda et al. [8] and Siirola [11] for the production of methyl-acetate from the reaction of methanol and acetic acid. Papalexandri and Pistikopoulos [12] proposed the idea of combination of phenomena to generate new unit operations. Peschel et al. [13], Lutze et al. [10, 14] and Babi et al. [2] illustrate the use of phenomena-based approaches to achieve process intensification.

Process integration can be considered as a special case of process intensification and is commonly defined as the design and analysis of the best (optimal) network for mass and energy utilization applied to the design (or retrofitting) and operation of new as well as existing processes [15]. Consequently, process integration is concerned with two integration concepts within a chemical (and biochemical) process—mass and/or energy integration [16, 17]. Mass integration is the efficient utilization of mass within the process (for example, minimization of fresh water use in a process) through the analysis and optimization of mass flows within the process. Analogously, energy integration is the efficient utilization of energy within the process (for example heat integration) through the analysis and optimization of energy needs within the process. As the use of water and energy are related to the performance criteria for more sustainable designs and are achieved through the integration of two or more operations, they are therefore also regarded as special cases of process intensification. Methods for performing process integration focus

on the selection and combination of existing as well as novel process technologies in the best (optimal) manner for the efficient use and utilization of mass and/or energy. Therefore, in this chapter, unless otherwise indicated, PI includes process intensification as well as process integration.

The objective of this chapter is to present and discuss the fundamentals of process intensification. First, the concept of PI is introduced, followed by a discussion on when and how to employ PI? This includes a discussion on the role of process intensification in sustainable process design as well as the connection between process integration and process intensification. Also, examples of successful application of process intensification are given. Next, the methods and tools needed to determine process intensification solutions are presented in terms of a general mathematical problem formulation, a brief overview of the different solution techniques, the need for a systematic computer-aided framework, and the associated methods and tools. The application of this framework is highlighted through a case study involving the production of methyl-acetate by reaction of methanol with acetic acid. The chapter ends with a discussion of perspectives, conclusions, and future work on the role of process intensification in relation to the design of more sustainable and innovative processes as well as pointing out some of the recent achievements. Note that even though process integration is regarded as a special case of process intensification, it is only briefly covered in this chapter. Also, recent developments on process integration can be found in the review of the state-of-the-art by Klemes et al. [18].

2.2 When and How to Apply PI?

In the chemical and biochemical process industry, improvements in economic as well as environmental factors are required for new as well as existing processes [3, 19]. PI plays a major role in achieving the desired improvements in processing options through the design of processes that constitute more sustainable alternatives, that is, hybrid/intensified unit operations (equipment). A hybrid/intensified unit operation, is an operation that enhances the function of one or more unit operations for performing a task or a set of tasks through a new design of the unit operation or the combination of more than one unit operations. For example, reactive distillation is a combination of reaction and separation (see Fig. 2.3a) and a membrane reactor is a combination of reaction and in situ removal of a reactant or product (see Fig. 2.3b).

Process Integration can be applied in the following forms for achieving process improvement:

- Heat integration (heat exchanger networks) where energy efficiency is increased through energy consumption minimization. This was the first type of process integration, first developed by Linnhoff et al. [20] and further extended and

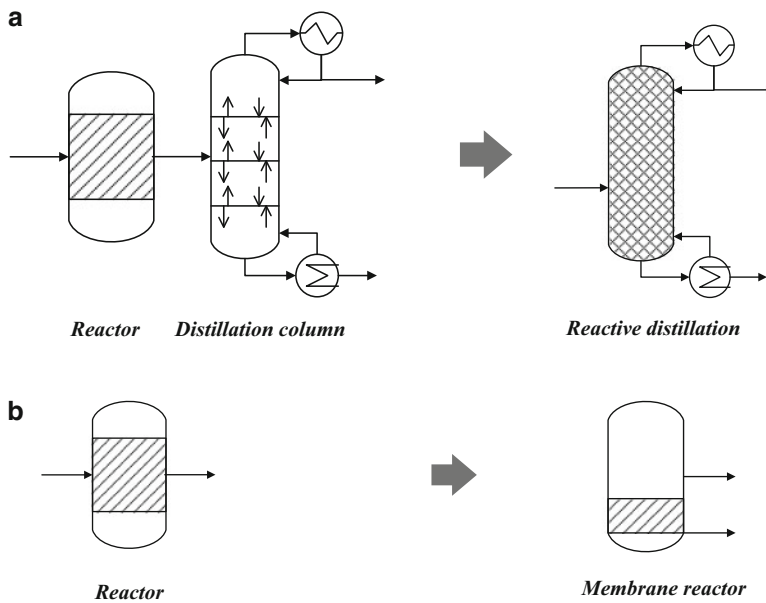


Fig. 2.3 Examples of hybrid/intensified unit operations: (a) reactive distillation (b) membrane reactor

solved using a mixed integer non-linear programming (MINLP) approach by Papoulias and Grossmann [21].

- Mass integration (mass exchanger networks) where the flow route of mass within the process is optimized, for example, through the use of concentration differences [16, 18].
- Supply-chain management where the total supply-chain cost (related to suppliers, storage, retailers, customers etc.), is minimized based on the concepts applied for heat and mass integration [18, 22].

The impact of process integration on cost reduction and environmental factors have been reported by numerous authors, see for example, Papoulias and Grossmann [21], Singhvi et al. [22] and Kazantzi and El-Halwagi [23] for overall capital cost and/or operating cost reduction through increased process integration. As the cost reduction due to more efficient use of energy and water resources is achieved, it also reduces the waste and has a positive impact on the environment, making the process thereby more sustainable.

Babi et al. [24] proposed a 3-stage sustainable process synthesis–design method (see Fig. 2.4) including the use of PI. According to this method, in the first stage, the optimal processing paths are synthesized to convert a set of raw materials into a desired set of products. In the second, the optimal processing path (flowsheet) is selected for further study that includes identification of the process bottlenecks or hot-spots. These hot-spots help to define targets for improvement, which when

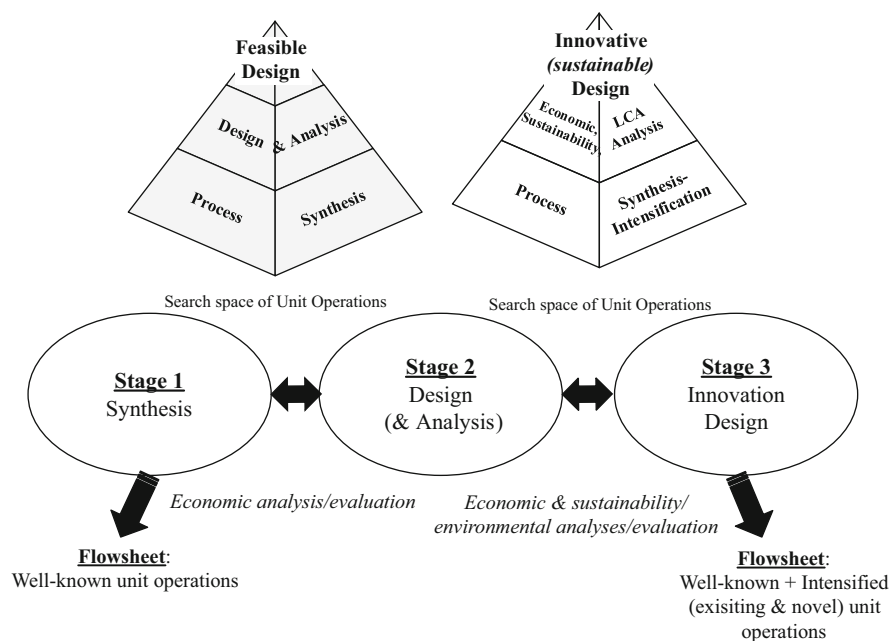


Fig. 2.4 Three-stage sustainable process design method that incorporates PI

matched would lead to more sustainable and innovative solutions. In the third-stage, methods with or without considering PI are applied to generate process alternatives that match the desired targets for process improvements. Here, applications of different PI methods are possible.

From Fig. 2.4, it can be noted that for existing processes with already identified hot-spots, entry to the 3-stage method is at stage 3. Alternatively, if the process exists but the hot-spots have not been identified, then the entry to the 3-stage method is at stage 2, while for a totally new process synthesis problem, the entry is at stage 1. Therefore, opportunities to apply PI exist whenever deficiencies (bottlenecks or hot-spots) in design and/or operation of a process are identified. A classic example is the flowsheet (see Fig. 2.5) for the production of methyl-acetate by reaction of methanol with acetic acid. Can the number of unit operations be decreased? Can the product yield and purity be increased? Can the process be made more sustainable? As Fig. 2.3b indicate, the answer is yes. Another classic example in the case of heat integration is the combination of a heat exchanger that needs to reduce the temperature of a stream with another that needs to increase the temperature of a stream. Integration in this case saves energy as well as cooling medium and thereby makes the process after integration more sustainable.

As the definition of PI implies, to apply PI, a combination of operations, tasks, and/or phenomena need to occur simultaneously subject to the specified objectives and constraints of the process. That is, which combination of operations, tasks,

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