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Introduction

1.1 Short History of Process Intensification

The timeline of process intensification (PI) (Figure 1.1) is about four decennia long. Although the term "process intensification" started to appear in East European publications on metallurgy already in mid-1960s and early 1970s, it was meant simply as equivalent to "process improvement." In chemical engineering, the first appearance of process intensification as we know it today was marked by the paper on application of centrifugal fields (so-called "HiGee") in distillation processes [1] published in 1983 by Colin Ramshaw from the ICI's New Science Group. The ICI project had been triggered by one of the NASA research projects on producing high transfer rates by using centrifugal fields in the zero gravity environment. Consequently, in the first years after its birth process, intensification was dominated by the rotating equipment, which still presents an important area of PI. Gradually, other technologies such as heat exchanger (HEX) reactors, intensive mixing devices, or microchannel reactors emerged within the PI domain.

Until early 1990s, process intensification was almost exclusively a British discipline. It was also the British BHR Group that organized the first international conference on PI in 1995 [2].

As can be seen in Figure 1.1, the real acceleration came in the last years of the second and, in particular, the first years of the third millennium, when a fast growth of PI-related activities in industry and in academia was observed. National academic–industrial PI networks have been established, first in the United Kingdom, later in the Netherlands, and in Germany. Process intensification has found its way to the university curricula. First books on PI were published [3–5] and the first PI-dedicated journal *Chemical Engineering and Processing: Process Intensification* was launched in 2007. In 2005, the European Federation of Chemical Engineering recognized the importance of PI by establishing the Working Party on Process Intensification (www.efce.eu/wp_pi). Since then, the Working Party organized five European Process Intensification Conferences (EPIC) and two International Process Intensification Conferences (IPIC) held in Barcelona in 2017 and in Leuven in 2019. The *European Roadmap for Process Intensification*, published in 2008 [6] and based on the contributions by the experts from 16 countries, laid down a foundation for short- and mid-term

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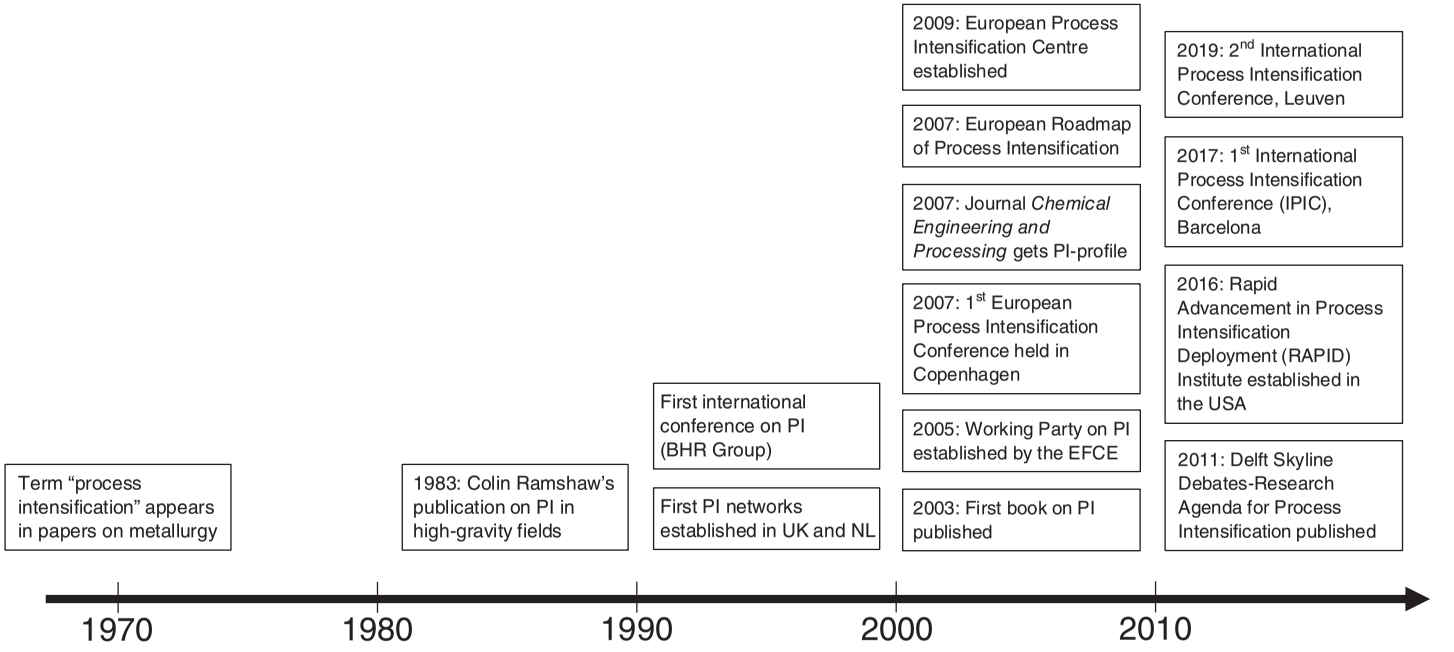

Figure 1.1 Timeline and milestones of process intensification.

research programs in the field. In 2009–2011, the roadmap got a follow-up in the form of the *Delft Skyline Debates* project, during which a multidisciplinary team of 75 leading academics and industrialists from different countries created a scientific vision on long-term developments in the field of process intensification that would reach beyond the horizon of 2050. The vision was published as a series of position papers [7] and also delivered a long-term research agenda for process intensification [8]. It is interesting to note that the above research agenda has gone beyond the traditional application area of PI, i.e. the chemical process industries, and has also addressed other areas including energy, water, and health. Also, in 2009, the European Process Intensification Centre (EUROPIC, www.europic-centre.eu) was established. The center presents an industry-driven platform for knowledge transfer in the field of PI and comprises chemical and pharmaceutical manufacturers, technology providers, equipment vendors, and engineering companies. Last but not least, the recently established *Rapid Advancement in Process Intensification Deployment (RAPID)* Institute (https://www.aiche.org/rapid), with private and governmental (US Department of Energy) funding exceeding 140 million dollars, presents a major development in this field in the United States and a proof of the importance of process intensification for the American economy.

It is interesting to note that after almost 40 years of development process, intensification has been brought again in connection with the space programs. Microchannel reactors for CO methanation are considered a promising technology for the *in-situ* resources utilization (ISRU) on Mars or on the Moon [9]. It appears that process intensification may one day revisit its birthplace – the Space.

1.2 Definitions and Interpretations of Process Intensification

From its very beginning, process intensification has been subject to numerous discussions and interpretations. In particular, various *definitions* of process intensification have been proposed in the literature, as presented in Table 1.1. Here, one can see that a considerable diversity exists in the way the researchers perceive and describe process intensification.

The *interpretations* of process intensification by various authors are quite diverse as well. For some, the *miniaturization* presents the fundamental issue of PI [22, 23], with microreactors being the common example. For others, process intensification is based on *functional integration* [14, 24], with reactive distillation as a prominent illustration. Arizmendi-Sánchez and Sharatt [25] combine different approaches by identifying *synergistic integration of process tasks and phenomena* and *targeted intensification of transport processes*, both on multiple scales, as the main PI principles. Finally, Freund and Sundmacher [26] present the suggestion that PI should and does follow a *function-oriented approach*. Similarly to the previous references, they identify different scales as to which this approach should be undertaken, from phase level via process unit level to plant level (thus not taking the molecular scale into account).

Table 1.1 Definitions of process intensification over the years.

Process intensification …	Author (year)	References
… [is the] devising exceedingly compact plant which reduces both the "main plant item" and the installations costs.	Ramshaw (1983)	[1]
… [is concerned with] order-of-magnitude reductions in process plant and equipment.	Heggs (1983)	[10]
… [is a] philosophy of plant design and construction whereby a given performance is achieved in very much smaller equipment – typically with a volume reduction of 2–3 orders of magnitude.	Ramshaw (1985)	[11]
... [is the] strategy of reducing the size of chemical plant needed to achieve a given production objective.	Cross and Ramshaw (1986)	[12]
…[is a] novel design approach where fundamental process needs and business considerations are analyzed and innovative process technologies used to meet these optimally.	Green (1998)	[13]
… [is the] development of innovative apparatuses and techniques that offer drastic improvements in chemical manufacturing and processing, substantially decreasing equipment volume, energy consumption, or waste formation, and ultimately leading to cheaper, safer, sustainable technologies.	Stankiewicz and Moulijn (2000)	[14]
… [is the] strategy of making dramatic reductions in the physical size of a chemical plant while achieving a given production objective.	Dautzenberg and Mukherjee (2001)	[15]
… [implies] faster reactions, better conversions, improved or new products and fewer by-products.	Swamy and Narayana (2001)	[16]
... [is the] revolutionary approach to process and plant design, development and implementation. Providing a chemical process with the precise environment it needs to flourish results in better products, and processes which are safer, cleaner, smaller, and cheaper.	BHR Group (2003)	[17]
… refers to technologies that replace large, expensive, energy-intensive equipment or process with ones that are smaller, less costly, more efficient or that combine multiple operations into fewer devices (or a single apparatus).	Tsouris and Porcelli (2003)	[18]
…[is] any chemical engineering development that leads to a substantially smaller, cleaner, safer and more energy-efficient technology.	Costello (2004)	[19]
... [is the] holistic approach starting with an analysis of economic constrains followed by the selection or development of a production process. Process intensification aims at drastic improvements of performance of a process, by rethinking the process as a whole. In particular, it can lead to the manufacture of new products which could not be produced by conventional process technology.	Degussa (Franke 2009)	[20]

(Continued)

Table 1.1 (Continued)

Process intensification …	Author (year)	References
… [provides] radically innovative principles ("paradigm shift") in process and equipment design which can benefit (often with more than a factor 2) process and chain efficiency, capital and operating expenses, quality, wastes, process safety and more.	European Roadmap for Process Intensification (2007)	[6]
… [stands for] an integrated approach for process and product innovation in chemical research and development, and chemical engineering in order to sustain profitability even in the presence of increasing uncertainties.	Becht et al. (2009)	[21]

Next to the diversity in definitions and interpretations of process intensification, the *relation between PI and Process Systems Engineering* (PSE) has also drawn a lot of attention and triggered many discussions within the chemical engineering community. Moulijn et al. [27] in a comparison between PI and PSE argued that PI had "a more creative than integrating character and primarily aims at higher efficiency of individual steps in that chain, for instance, by offering new mechanisms, materials, and structural building blocks for process synthesis." They also saw process intensification acting more on the nanoscale (molecules) to macroscale (reactors) and less on the megascale (plants, sites, and enterprises). We share both these views. Process intensification should indeed be seen as a "supplier" of novel building blocks, from which process systems are synthesized, as shown in Figure 1.2.

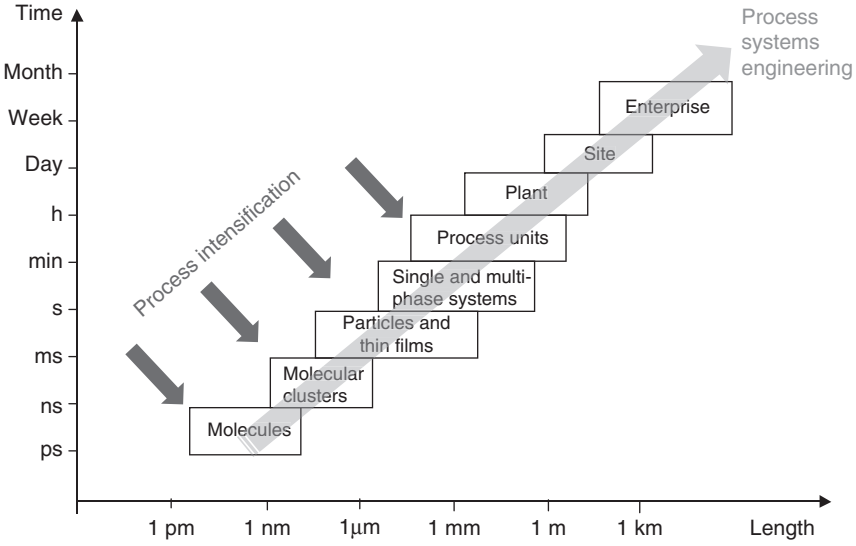

Figure 1.2 Process intensification delivers new concepts of individual steps in Process Systems Engineering chain, at small and medium scales (molecules to process units). Source: Adapted from Grossmann and Westerberg 2000 [28].

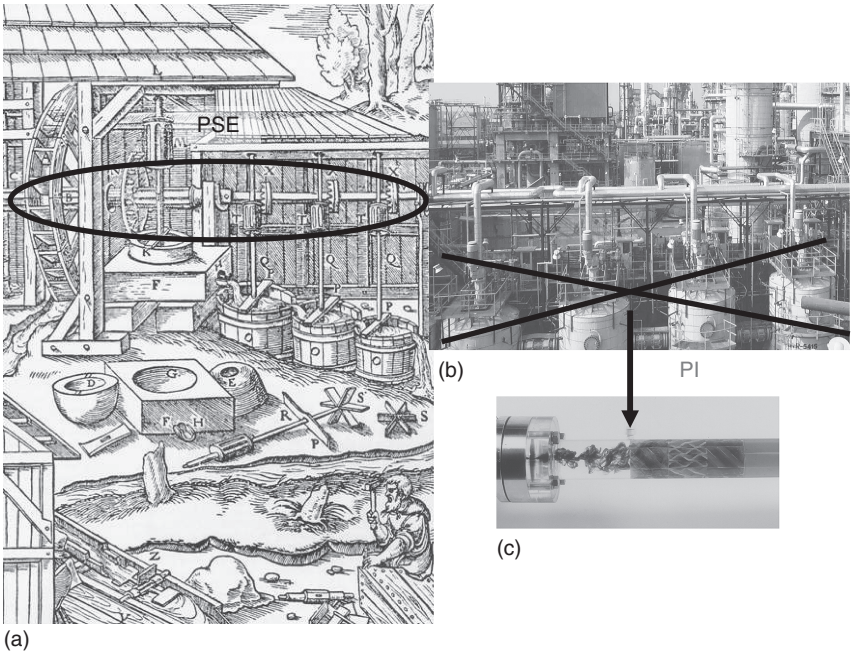

Figure 1.3 The relationship between PSE and PI: PSE integrates the individual steps, whereas PI introduces new concepts of those steps. Source: (a) Agricola 1556 [29]; (b) Stankiewicz 2006 [30]; (c) Sulzer Chemtech Ltd., Switzerland, www.sulzer.com.

Moulijn et al. [27] illustrated the distinction between PI and PSE with old and modern chemical plants shown in Figure 1.3. The PSE thinking in Middle Ages is presented in the left-hand side figure that comes from the sixteenth century book "De Re Metallica" by Georgius Agricola [29]. It illustrates the process of retrieving gold from ore. Ore is crushed by the stamp "C," ground in the mill "F," and mixed with mercury in vessels "O." Remarkably, these three operations are integrated and driven by a water wheel via a shaft and a number of gears. This is an example *par excellence* of the PSE thinking. On the other hand, a motionless mixer as a replacement of the stirred tanks in the modern plant presents a much more efficient, fundamentally different concept of mixing fluids and a classical example of process intensification.

1.3 Fundamentals of Process Intensification – Principles, Approaches, Domains, and Scales

In 2000, Stankiewicz and Moulijn [14] proposed a "toolbox" view of process intensification and divided that "toolbox" into two subdomains (Figure 1.4):

- *Process-intensifying equipment*: such as novel reactors, intensive mixing, heat transfer, and mass transfer devices;

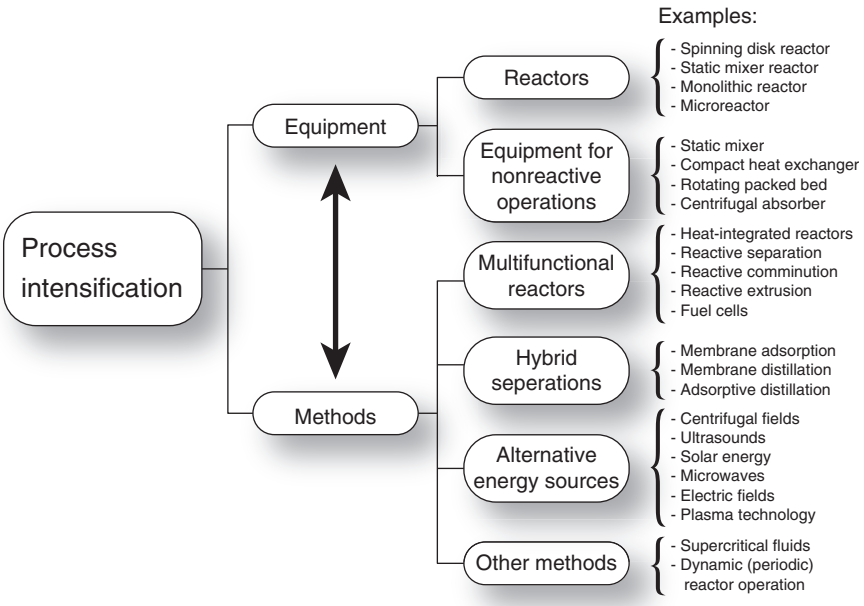

Figure 1.4 Process intensification as a "toolbox" with equipment and methods. Source: Adapted from Stankiewicz and Moulijn 2000 [14].

- *Process-intensifying methods*: such as new or hybrid separations, integration of reaction and separation and/or heat exchange and/or phase transition (in so-called multifunctional reactors), techniques using alternative energy sources (light, ultrasound, etc.), and new process control methods (e.g. intentional unsteady-state operation).

Obviously, in some cases, overlaps between these two domains can be observed as new methods often require novel types of equipment to be developed and vice versa, novel apparatuses already developed make sometimes use of new, unconventional processing methods.

The above-described classical "toolbox" view had functioned for almost a decade and was adopted or referred to in numerous publications. However, in the course of time, it became apparent that the "toolbox/examples" approach to process intensification was insufficient for a comprehensive analysis of existing and systematic development of new, intensified processes. It could be used for an incidental improvement of one or another process step or equipment, but it lacked the generic dimension, which is needed for a multiscale, holistic process design and development. To allow the latter, process intensification needs to be approached and understood in a more fundamental way. This is also the primary aim of the present book.

We will start the book by analyzing *the generic principles of PI*. Earlier approaches (e.g. [31]) defined the ultimate goal of process intensification as achieving a purely kinetics-limited process without any limitations resulting

from the momentum, heat, and mass transfer. This is surely a valid approach, but it addresses only a part of the problem as, as you will read further in this book, reaction kinetics can also be intensified. By saying "intensified," we do not mean operating at higher temperatures or using catalysts. Fundamental research in the field of chemical physics shows that one can boost the kinetics of a chemical reaction by orders of magnitude providing the right form of energy to chemical bonds, without using a catalyst and without increasing the macroscopic temperature even by one degree! This proves that kinetic limitation is indeed a relative concept. Furthermore, the earlier approaches to process intensification neglected two other elements that are of great importance for an intensified process, namely the uniformity of the processing history for all molecules and the synergistic effects resulting from interactions and interrelations between various process steps at different scales.

In view of the above considerations, an ideal intensified processing system (and the *ultimate goal of PI*) is a system in which reactions proceed at a maximum achievable efficiency, all molecules undergo the same processing history, the hydrodynamic, heat and mass transfer limitations are removed, and the synergies resulting from interrelations between various operations and steps are fully utilized. Such a definition of the ultimate goal of PI leads us directly to the following four guiding principles, which we call *generic principles of process intensification*:

- Maximize the effectiveness of intra- and intermolecular events;
- Give each molecule the same processing experience;
- Optimize the driving forces and resistances at every scale and maximize the specific surface areas to which these forces or resistances apply;
- Maximize the synergistic effects from partial processes.

The above principles, in one form or another, are obviously not entirely new to chemical engineering. In process intensification, however, they result from the explicit definition of the goal that an intensified process aims to reach. Besides, the PI interpretation of these principles often goes beyond the boundaries of the classical chemical engineering. This can be seen, for instance, in the first principle, where process intensification looks at the molecular-scale methods for improving the intrinsic kinetics of chemical reactions, such as molecular alignment, orientation, and selective bond excitation.

In the realization of the above guiding principles, process intensification operates in *four elementary domains*: spatial, thermodynamic, functional, and temporal (Figure 1.5). A detailed analysis and discussion of those four domains and the corresponding PI approaches constitute the main part of this book.

Finally, one needs to stress that the above-described principles and approaches should address all *relevant time and length scales* (Figure 1.5). As you will see further in the book, this is indeed the case. Looking more into the future, we believe that, whereas process intensification until recent years has mostly been focused on the larger scales (from films and particles up to processing units), the major breakthroughs to come will arise from process intensification on the molecular scale.

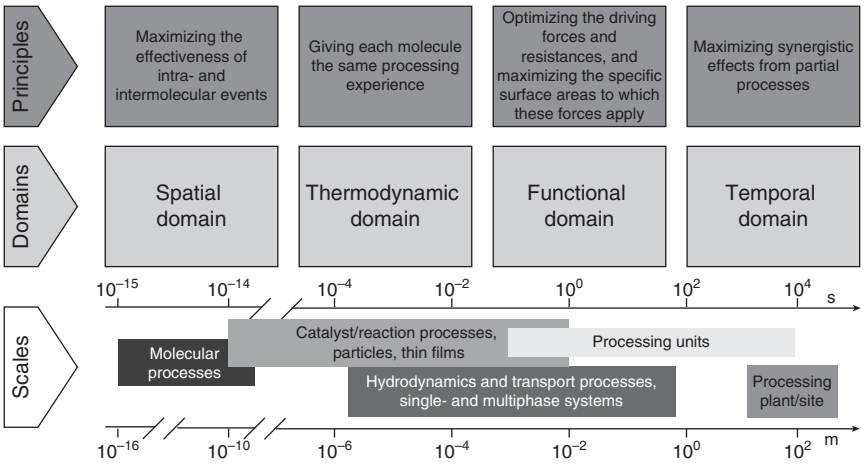

Figure 1.5 Principles, approaches, and scales of process intensification. Source: Adapted from Van Gerven and Stankiewicz 2009 [32].

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