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Computers and Chemical Engineering

journal homepage: www.elsevier.com/locate/compchemeng

From process integration to process intensification *

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ARTICLE INFO

Article history: Received 5 December 2014 Received in revised form 19 February 2015 Accepted 19 March 2015 Available online 30 March 2015

Keywords: Process intensification Process integration Process synthesis Process control

ABSTRACT

In this paper, we establish a connection between process integration and process intensification. Focusing on processes with material recycle, we use an asymptotic analysis to demonstrate that intensification represents a limit case of tight integration through significant material recycling. Based on this result, we propose a novel avenue for discovering intensification opportunities at the process design stage. Subsequently, we investigate the dynamics and control implications of the transition from process integration to process intensification. We demonstrate that, for the same steady-state performance, the dynamic response of an integrated process is slower than that of its intensified equivalent. Also, we provide a theoretical justification for existing empirical arguments concerning the loss of control degrees of freedom caused by process intensification. The theoretical developments are applied on a reaction–separation–recycle process example.

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1. Introduction

In its age of maturity in the second half of the 20th century, the chemical industry has favored increasingly larger plant designs, whose scale led to lower production costs for bulk chemicals and petrochemical products. Starting in the 1970s, scarcer and more expensive energy resources, and tighter environmental regulation have spurred process integration efforts. Integrated processes rely on material recycling to minimize raw material use and emissions, and make extensive use of heat recovery to improve energy efficiency. Such integration measures are currently implemented in most chemical and petrochemical production facilities worldwide (Baldea and Daoutidis, 2012).

Process intensification takes a different perspective on maximizing the efficiency of a process, focusing on optimizing driving forces (Van Gerven and Stankiewicz, 2009) such that process reactions are governed by their intrinsic rates rather than by transport/transfer phenomena. This is typically accomplished by operating at space scales that are smaller than in conventional processes, under the assumption that scale-up can be accomplished by increasing the number of intensified units that operate in parallel (Reay et al., 2013).

http://dx.doi.org/10.1016/j.compchemeng.2015.03.011 0098-1354/© 2015 Elsevier Ltd. All rights reserved. Based on these considerations, we can compile a definition of process intensification as:

Any chemical engineering development that leads to substantially smaller, cleaner, safer and more energy efficient technology (Reay et al., 2013) or that combine[s] multiple operations into fewer devices (or a single apparatus) (Tsouris and Porcelli, 2003).

It is thus to be expected that the capital costs, and physical and environmental footprints of intensified processes are lowered compared to their conventional, non-intensified counterparts. Moreover, smaller physical dimensions can be adjusted to ensure that every molecule of material can experience the same processing conditions (Van Gerven and Stankiewicz, 2009).

Many interesting process intensification applications focus indeed on the small scale. These include, e.g., miniaturized fuel processing systems (Kolios et al., 2005), miniaturized (Karim et al., 2008) and portable power sources (Yunt et al., 2008), high-throughput experimentation and reaction development (McMullen and Jensen, 2010).

The "multum in parvo" ("much in little") intensification concepts have also been successfully implemented at the industrial scale. Of the intensified versions of the core reactor–separator structure, *reactive distillation* has been hailed as "the front runner of industrial process intensification" (Harmsen, 2007); *membrane reactors* have found applications in high temperature gas separations (De Falco et al., 2011; Anon., 2012). Dividing wall columns which combine multiple distillation towers in a single physical device (Asprion and Kaibel, 2010), were shown to lead to significantly lower capital and operating costs for separation processes (Schultz et al., 2002).

[†] A preliminary version of this work was presented at Foundations of Computer-Aided Process Design (FOCAPD) 2014, Cle Elum, WA (Baldea, 2014).

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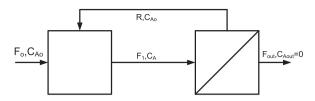


Fig. 1. Integrated reactor-separator-recycle process (Baldea and Daoutidis, 2014).

Catalytic plate reactors are used in compact plants for hydrocarbon processing (e.g., steam methane reforming (Zanfir et al., 2011; Pattison and Baldea, 2013; Pattison et al., 2014), Fischer-Tropsch synthesis (Roberts, 2013; LeViness et al., 2014)).

These developments are mainly the result of relentless experimentation by practitioners rather than the outcome of insights derived from rigorous theory. Indeed, process intensification appears to be one of the fields where practice is – often by a good distance – ahead of theory. The development of a systematic approach for analysis and discovery of intensified process alternatives during process synthesis is in its incipient stages.

Devised in the first half of the 20th century, the unit operations paradigm has dominated process synthesis in the past decades. Its advent and longevity are well motivated: unit operations provide a consistent approach to building process functionality that meets a desired outcome in terms of production rate and product quality. Moreover, unit operations facilitated the development of meaningful and fast shortcut design calculations in an era when computational resources were scarce and costly¹. Recent efforts in process intensification (e.g., Arizmendi-Sánchez and Sharratt, 2008; Lutze et al., 2013) advocate abandoning the unit-operations framework in favor of representing a process as a set of phenomena (Papalexandri and Pistikopoulos, 1996), which serve as potential building blocks for intensified devices.

In this paper, we propose a different approach for identifying intensification candidates at the flowsheet level. Focusing on integrated processes with material recycling, we establish a connection between process integration and process intensification. Specifically, we use an asymptotic analysis to demonstrate that intensification represents a limit case of tight integration through significant material recycling. Moreover, we investigate the process control implications of the transition from process integration to process intensification. We demonstrate that, for the same steady-state performance, the dynamic response of an integrated process is slower than that of its intensified equivalent. Also, we propose a theoretical justification for existing empirical arguments concerning the loss of control degrees of freedom caused by process intensification. Finally, the theoretical developments are applied on a reaction-rate-based-separation-recycle process example.

2. Introductory example

To begin exploring the connection between process integration and process intensification, we propose studying a "conventional" process comprising distinct unit operations. The reaction-separation-recycle is perhaps the most pervasive structure (Baldea and Daoutidis, 2012) for such systems, and is illustrated in the following simple example.

Consider the case of a CSTR followed by an ideal separator (Fig. 1). Reactant is converted to product in a first-order reaction. The effluent undergoes separation in an ideal separator, after which

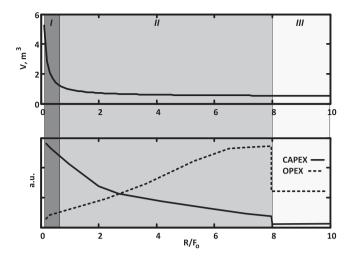


Fig. 2. (top) Reactor volume as a function of recycle rate in the conventional (I), integrated (II) and intensified (III) regions. (bottom) Empirical estimate of capital and operating costs (CAPEX, OPEX) for the three regions.

a stream of pure, unreacted material is returned to the reactor. For simplicity, the operation is assumed to be isothermal. The design and operation of this process is driven by the reaction kinetics; thus, a slow reaction must be compensated for by increasing the reactor volume and/or increasing the recycling rate.

The connection between these design variables can be easily captured by the steady-state component balance equations:

$$0 = (F_o + R)C_{Ao} - F_1C_A - kC_AV$$

$$0 = F_1C_A - RC_{Ao}$$
(1)

where *V* is the reactor volume and *k* is the reaction rate constant. The plot at the top of Fig. 2 illustrates the relationship between reactor size and recycle rate R for the specific case where $k = 0.01 \text{ s}^{-1}$, $C_{Ao} = 1000 \text{ mol}/m^3$, $F_o = 0.01 \text{ m}^3/s$. Based on the value of the recycle flow rate and the reactor volume, one can distinguish three separate design regions. Low values of R (region I) call for a large reactor volume. This is the case of a conventional process system, which has the highest capital cost (CAPEX) relative to the options that we elaborate on in the sequel, as illustrated in Fig. 2 (bottom). Process integration (region II) relies on a significant increase of material recovery and recycling to improve process performance and reduce equipment size (Baldea and Daoutidis, 2012). Thus, reactor size (and, correspondingly, the CAPEX) of an integrated process is lower, at the price of a higher operating expense (OPEX) due to increased costs associated with recirculating the material between the reactor and the separator. Finally, region III corresponds to very high material recycling rates. Here, interactions between the reaction and separation units are very strong (see, e.g., Kumar and Daoutidis, 2002; Baldea and Daoutidis, 2014, 2012) and the two units can be construed as acting as a single entity from a dynamic point of view. Region III can thus be regarded as the process intensification regime, in which reaction and separation occur in a single, combined device. Assuming that such a device can be built, operating costs are likely to drop dramatically owing to the complete elimination of recycling. Likewise, eliminating one unit will yield a CAPEX reduction.

Based on the analysis above (carried out on an admittedly simple model), it can be inferred that process intensification offers significant potential benefits from a design point of view, including reducing the number of units and the size of the required equipment and eliminating the capital and operating cost associated with recycling.

¹ The author is grateful to Prof. James B. Rawlings and Dr. Robert Turney for drawing his attention to these points.

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