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Chemical Engineering Research and Design

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


Process intensification from conventional to advanced distillations: Past, present, and future

Zong Yang Kong^a, Eduardo Sánchez-Ramírez^b, Ao Yang^c,
Weifeng Shen^{d,*}, Juan Gabriel Segovia-Hernández^{b,*}, Jaka Sunarso^a

^a Research Centre for Sustainable Technologies, Faculty of Engineering, Computing and Science, Swinburne University of Technology, Jalan Simpang Tiga, 93350 Kuching, Sarawak, Malaysia

^b Universidad de Guanajuato, Campus Guanajuato, División de Ciencias Naturales y Exactas, Departamento de Ingeniería Química, Noria Alta s/n, 36050 Guanajuato, Gto, Mexico

^c College of Safety Engineering, Chongqing University of Science & Technology, Chongqing 401331, PR China

^d School of Chemistry and Chemical Engineering, Chongqing University, Chongqing 400044, PR China

ARTICLE INFO

Article history:

Received 12 September 2022

Received in revised form

27 September 2022

Accepted 29 September 2022

Available online 3 October 2022

Keywords:

Process intensification

Advanced distillation

Reactive distillation

Extractive distillation

Reactive-extractive distillation

Sustainability

ABSTRACT

This perspective paper features the process intensification (PI) application for advanced distillation-based processes. Starting with the historical background of generic PI, we subsequently narrow down the discussion to extractive distillation (ED), reactive distillation (RD), and hybrid reactive-extractive distillation (RED). We categorize the existing PI techniques onto internal and external intensification, where the former does not involve altering the distillation configuration while the latter does. Instead of deliberating the technical aspects, we explicitly highlight the contribution of PI applied to ED, RD, and RED towards societal impact covering energy, economic, environmental, control, and safety perspectives. The future perspectives of PI are discussed in the last section, covering the development of hybrid PI technologies, exploring the energy efficiency of different PI configurations, prioritizing PI beyond energy by considering some other sustainability aspects, and linking PI with the ever-increasing Industry 4.0 applications.

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1. Introduction

With an increasing awareness of sustainability issues and a highly competitive global market, chemical production is making its way towards a paradigm shift to achieve more

efficient, cost-effective, environmentally friendly, safe, and versatile production. Process Intensification (PI) aims to increase energy efficiency and process safety, and improve economic profitability, while reducing waste and emissions by adopting innovative designs, which may be out of the

Abbreviation: ABE, acetone, butanol, and ethanol; AI, artificial intelligence; CPS, cyber-physical systems; CC-I4.0, convergence of I4.0; DL, Deep Learning; DEG, diethylene glycol; DCRED, double-column reactive-extractive distillation; DW-RED, dividing wall reactive-extractive distillation; DWC, divided wall column; EGMA, ethylene glycol monoacetate; EGDA, ethylene glycol diacetate; ESI, ease of separation index; ED, extractive distillation; EI99, eco-indicator 99; EO, ethylene oxide; EG, ethylene glycol; GHG, greenhouse gas; IR, individual risk; I4.0, Industry 4.0; IoT, Internet of Things; ML, Machine Learning; ORC, organic rankine cycle; PSD, pressure swing distillation; PI, process intensification; PC, preconcentration column; QM, Quantum Mechanical; RD, reactive distillation; RED, reactive-extractive distillation; SVD, singular value decomposition; SRC, solvent recovery column; SS, side-stream; TCD, thermally coupled distillation; TAC, total annual cost; TCRED, triple column reactive extractive distillation; UN, United Nation; UN-SDGs, UN Sustainable Development Goals; VRC, vapor recompression

* Corresponding authors.

E-mail addresses: zkong@swinburne.edu.my (Z.Y. Kong), shenweifeng@cqu.edu.cn (W. Shen),

gsegovia@ugto.mx (J.G. Segovia-Hernández), jsunarso@swinburne.edu.my (J. Sunarso).

<https://doi.org/10.1016/j.cherd.2022.09.056>

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vision of the current industry practice (Pistikopoulos et al., 2021). The idea of PI was first introduced to the chemical industry in the 1970s and has manifested into some of the processes and equipment innovations widely used to this day, for example, static mixer and reactive distillation. In recent decades, PI has attracted increasing academic and industrial interests as a guide for process improvement in the design of new facilities, as well as for the modernization of the existing ones to meet the increasing demands for sustainable chemical production. Ramshaw (1995) presented at the “1st International Conference on Process Intensification for the Chemical Industry” in 1995, one of the first definitions of PI: “a strategy to reduce the size of a chemical plant by reducing the number of unit operations and parts of team involved”. Such definition was somewhat limited, as it refers exclusively to downsizing plant and/or equipment. Thus, Stankiewicz and Moulijn (2000) redefined the scope of PI as “the development of new equipment and processes that significantly reduce the size of equipment and plants, reduce waste production, which results into more economic and sustainable production”. Ten years ago, Ponce-Ortega et al. (2012) and Reay et al. (2013) further expanded the definition of PI to include principles such as increased energy efficiency, production yield, process safety, and reduced use of raw materials and inventories. Over the last century, chemical engineers have developed modern technologies to produce various agricultural and basic chemicals, fuels, fertilizers, pharmaceuticals, and materials that facilitated economic growth in industrialized countries. While chemical engineering is expected to contribute to United Nation’s (UN) goals, applying traditional process design and optimization strategies will not bring about these changes fast enough. In addition, modern chemical industrial processes are carried out in large integrated chemical industries with a limited degree of freedom to shift from fossil-based feedstocks and energy carriers to renewable resources (Bharat Petroleum Corporation Limited, 2018). A paradigm shifts on energy sources, raw materials, and the scale of the chemical industry is required to achieve the UN Sustainable Development Goals (UN-SDGs). In this context, PI strategies offer changes in the raw material/energy transition and process efficiency (Rivas et al., 2018). This potential has been recognized by legislators, technology providers, and end users. In 2006, for example, SenterNovem, an agency of the Netherlands Ministry of Economic Affairs, which implements programs based on innovation and sustainability on behalf of the government, defined the benefits of PI as follows: (1) energy saving in the range of 20–80%, (2) capital and operating expenses saving of 20–80%, (3) chemical inventory reduction of 10–1000 times, and (4) significant improvement in yield and selectivity transition (Rivas et al., 2018).

In general, the literature has summarized at least seven activities (Segovia-Hernández and Bonilla-Petriciolet, 2016), which will result in intensified processes: (1) combine multiple tasks or process equipment into a single unit (e.g., reactive distillation (RD)), (2) tight process integration (e.g., integration of materials or energy), (3) use of new multifunctional materials (e.g., ionic liquid or deep eutectic liquids), (4) miniaturization of process equipment (e.g., microreactor), (5) switching mode of operation (e.g., pressure swing adsorption or distillation), (6) application of improved driving forces (e.g., rotating packed bed), and (7) advanced operating strategies (i.e., advanced control). Current efforts towards PI through process systems engineering approaches

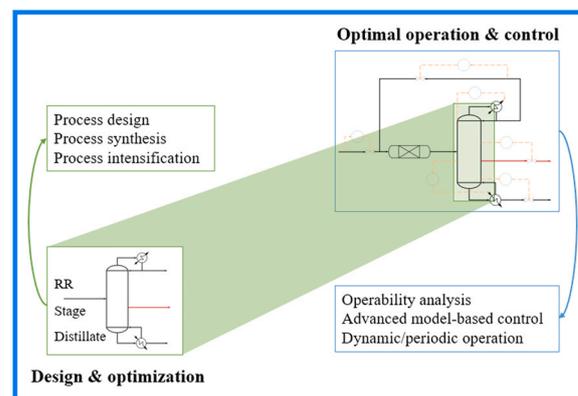


Fig. 1 – Classification of current efforts toward PI through process system engineering approaches.

can be broadly classified based on (a) design and optimization and (b) optimal operation and control (Pistikopoulos et al., 2021), which are graphically illustrated in Fig. 1. Today, the reverse flow reactor, RD and thermally coupled distillation (TCD) sequences, to name a few, are some of the examples of successful PI application that are already commercialized and are examples of PI’s technological developments.

Distillation is one of the oldest unit operations in the chemical industry, which remains as the workhorse of separations, widely used for its simplicity and efficiency (Sholl and Lively, 2016). It is present in some of the largest and most profitable separations in the world, such as hydrocarbon separation, crude oil fractionation, cracking, and natural gas liquid separation. Within chemical process plants, distillation accounts for 90–95% of all separation processes and consumes 40–60% of the energy in the chemical and refining industries. However, it is well known that the distillation process has several drawbacks such as the high capital and operating costs. The thermodynamic efficiency of distillation is also low (e.g., about 12% for crude oil separation, about 18% for air separation, and about 5% for C2 and C3 separation) (Kiss and Smith, 2020). Another key problem is that distillation uses energy of high cost and quality (usually steam coming from reboilers), which is eliminated in the condenser at low temperature and, therefore, a thermodynamic problem of exergy loss is generated. For this reason, achieving greater efficiency and lowering the operational cost are crucial points to stay competitive in the chemical process industry. Although there are uses of great industrial relevance for other separation processes, such as adsorption, membranes, liquid-liquid extraction, and absorption, distillation has key advantages over all those alternatives. For example, distillation can separate mixtures with a wide range of feed concentrations and produce products of high purity, and it can also provide a wide range of yields (from very low to very high). All these advantages will keep distillation as the key separation in process industries for years to come. If the distillation process will continue to be a key operation in the chemical industry and a large consumer of energy (generating greenhouse gas emissions (GHG) when operating with fossil fuels), work must be done on alternatives and proposals to reduce energy consumption in distillation (Kiss and Smith, 2020).

In the context of multicomponent distillation, PI is believed to represent innovative process synthesis strategies that minimize the amount of equipment, increase the energy

efficiency of multicomponent distillation systems, and reduce the total operation cost (Jiang and Agrawal, 2019). Gómez-Castro and Segovia-Hernández (2019) commented that among the PI technologies proposed to improve distillation, the most promising are in the functional domain (synergy), since they integrate various unit operations in a single unit, thus taking advantage of synergistic effects to overcome equilibrium limitations. This allows the design of compact equipment with greater thermodynamic efficiency. Hybrid technologies based on process intensification principles lead to more efficient integrated distillation processes. Such is the case with Divided Wall Columns (DWC), TCD, RD, Cyclic Catalytic Distillation, and many others.

This perspective article focuses on the PI from simple to advanced distillation-based processes, particularly on extractive distillation (ED), RD, and reactive-extractive distillation (RED). Although there are already a few review papers that discuss these advanced distillation-based processes such as ED (Lei et al., 2003), AD (Li et al., 2005), PSD (Mahdi et al., 2015; Shen et al., 2016), these reviews are mostly technical-based that cover the selection of solvents, mathematical models, and thermodynamic analyses. Some of these reviews also summarize the recent studies and point out the gaps as part of the recommendation for future work. In comparison to previous reviews, this article aims to provide the authors' knowledge and perspective on the PI applied to advanced distillation-based processes, particularly on ED, RD, and RED. Generally, these processes (e.g., RD and ED) are far more industrially established in comparison to the other advanced distillation-based process such as membrane distillation (Gerbaud et al., 2019; Kiss, 2013; Ma et al., 2019; Sharma and Mahajani, 2002). Starting with the historical application of PI in simple distillation in Section 1, we deliberately shifted the discussion to advanced distillation-based process. In Section 2, we categorized the existing PI techniques into external and internal intensifications and summarizes their features to provide the readers with sufficient fundamentals. Then, the current application of PI to the advanced distillation-based processes are outlined in Section 3, together with the authors' opinion on the contribution of PI towards societal impact covering energy, economic, environmental, control, and safety aspects. The future perspective of PI towards ED, RD, and RED applications are overviewed in Section 4 while Section 5 concludes this perspective paper.

2. Types of intensifications

The overall goal of PI is to synthesize compact, easy-to-operate, energy efficient, and cost-effective configurations for a given separation task. Here, it is discussed two important strategies for achieving this goal. The first strategy involved those techniques which require additional auxiliary equipment but does not usually alter the distillation configuration. The second strategy deals with internal intensification. This technique usually involved altering the distillation configuration such as removing one unit of reboiler (e.g., TCD or DWC) or to install an additional side-stream (SS).

2.1. External intensification

Following the oil crisis of the 1970s, there was a resurgence of the interest in thermal intensification. The main energy consumers in separation processes were refrigerant-cooled

compressors, reboilers, and condensers. Thermally integrated fractionation machines incurred higher equipment cost but were compensated by lower utility costs. However, due to rising energy costs that rose faster than the equipment costs and the environmental concern brought on by GHG emissions, the heat integration (HI) strategy has attracted a lot of research interest in the literature and is more economically viable for distillation operations.

Among the several HI distillation systems, the heat pumping system has become one of the continuously endorsed designs for continuous flow distillation columns (Yang et al., 2019c). Studies conducted in actual industrial application have shown that this system could dramatically reduce the net energy consumption and GHG emissions. In a conventional distillation column, heat is produced at the bottom reboiler supplied by a hot utility and is lost to the top condenser by a cold utility, resulting in a large energy consumption. From the thermodynamic perspective, the integration of energy between condenser and reboiler, which represent the main source and sink of energy, respectively, are a clear way to reduce the dependence on the external heating utilities. The connection of a heat pump and a distillation column is referred to in this sentence as "integration.". In mechanical heat pump, the vapor stream that leaves the top tray is compressed to a higher pressure and then utilized to heat the bottom liquid.

The mechanical heat pumps are electrically powered vapor recompression (VRC) system. Early in the 1950s, Freshwater (1951) popularized the concept, and Null (1976) has subsequently created three essential approaches based on this concept: bottom flashing, external VRC, and direct VRC. In contrast to the other two mechanical techniques, Henley and Seader (1981) reported the use of a heat pump with reboiler liquid flashing for the separation of a propylene-propane (P-P) combination and demonstrated its highest thermal efficiency. They demonstrated that employing a combination of CFC-12 and CFC-114 as the working fluid boosts energy efficiency in a closed cycle system by up to 30–75% when top condenser and bottom reboiler were added to intermediate heat exchangers. A laboratory-scale VRC column for separating a binary combination of ethanol and water was devised and constructed by Canales and Marquez (1992) and they found that a reduction of energy consumption between 45% and 56% can be realized in comparison to the conventional column. Díez et al. (2009) evaluated the direct VRC, bottom flashing, and absorption heat pumps using *i*-butane/*n*-butane combination and demonstrated through simulated studies that distillation with top VRC and bottom flashing configurations both enable reductions in capital costs of 9% and 10% and operational costs of 33% and 32%, respectively. Kiss et al. (2012) have suggested a set of rules for choosing the most promising thermally integrated distillation column: different boiling points, the type of separation task, product flow and parameters, operating pressure, reboiler duty, and temperature. These rules have been the focus of extensive research, with particular attention paid to design and optimization (Ferre et al., 1985; Fitzmorris and Mah, 1980), modeling (Brousse et al., 1985; Oliveira et al., 2001), operability analysis (Annakou and Mizsey, 1995; Enweremadu et al., 2009), and control (Hernandez, 1981; Jogwar et al., 2010; Muhrer et al., 1990).

On the other hand, the second industrial revolution was mostly fueled by the advent of electricity, and the steam Rankine cycle, which is now the dominant method of

producing electricity, is still driven by fossil fuels. Several thermodynamic cycles, including the organic Rankine cycle (ORC), have been suggested and studied for the conversion of low-grade heat into electricity (Chen et al., 2010). Recent years have seen a rapid increase in the number of researches on the use of ORC for producing power from waste heat (Yang et al., 2019b). The ORC's working fluid is an organic medium, where the liquid-vapor phase transition occurs at a lower temperature than the water-steam phase transition, but the Rankine cycle's basic principles still apply. To lower environmental pollution, ORC technology adoption is essential. It uses heat with a low supply temperature and effectively utilizes waste heat generated by industrial systems. Yu et al. (2018) noted that one of the most crucial factors depends on the characteristics of the heat source, which relies on the selection of an appropriate working fluid due to the poor efficacy of ORC. Using appropriate working fluid can increase the ORC's thermal efficiency. The designs of the manufacturer, the installed apparatus, the working fluid, and the fluctuating temperature of the waste heat supply are also important parameters. Depending on the working conditions, the operational parameters of ORC may function differently and have different values. Generally, the conversion efficiency ranges from 10–20%.

2.2. Internal intensification

Apart from external intensification, another alternative strategy is to intensify the distillation process by making changes to the topology of the system (Contreras-Zarazúa et al., 2021). For example, this can be performed by modifying the conventional distillation configuration through removing a reboiler unit to generate a TCD. Separation of multi-component mixtures into three or more products is typically accomplished by conventional distillation column sequences with one feed stream and two product streams with one reboiler and one condenser per column. Some complex distillation arrangements, with or without thermal coupling (and decreased number of reboilers and condensers), have been shown to provide significant energy and exergy savings. Such incentives have aroused considerable scientific interest in the last 20 years in the area of distillation column topology modification. A good number of works on the design and analysis of TCDS for ternary separations have been reported (i.e., the classic paper by Malone (1988), Tedder and Rudd (1978) or the recent study by Sun et al. (2020)). These studies have shown that the thermally coupled configurations can achieve energy savings of up to 30% in contrast to conventional direct and indirect distillation sequences for the separation of mixtures with low or high content of the intermediate component. For ternary, quaternary, and multi-component zeotropic mixtures, various arrangements for TCD configurations have been proposed. Despite the potential benefits of thermally coupled columns and some reports of successful industrial applications, only a limited number of such columns have been implemented. The TCD has been largely applied in Europe and Japan given their dependence on imported crude oil (Contreras-Zarazúa et al., 2021).

The term of DWC is defined as dividing wall column where the middle part of the column is split into two parts by a wall. Feed, typically contains three or more components (A, B, and C), is pumped into one side of the column facing the wall. Even though this concept was established some 60

years ago, only in 1985, BASF developed the first industrial DWC into operation (Segovia-Hernández et al., 2021). BASF also started up the first 4 products DWC. Theoretical studies have shown that DWC (for ternary and multicomponent mixtures) can save up to 30% in the capital invested and up to 40% in energy costs (Segovia-Hernández et al., 2021). Moreover, retrofitting conventional column systems into DWC is also a feasible option if pressure conditions and design specifications are met. Several other successful examples of use of DWC can be found among reactive separations that combine reaction and separation steps in a single unit, such as RD (Kiss and Smith, 2020). To promote its industrial implementation, a proper understanding of the control properties and operation of the DWC is needed to complement the energy savings results (Contreras-Zarazúa et al., 2021). Evidently, the anticipation that the dynamic properties of a complex distillation column may cause control difficulties compared to the rather well-known behavior of the conventional direct and indirect sequences for the separation of ternary mixtures may be one of the factors that had contributed to their lack of industrial implementation (Vallejo-Blancas et al., 2022).

Instead of using the thermal couplings as in the cases of TCD and DWC, another attractive alternative PI is the use the liquid only SS, commonly known as the SS configuration. Such configuration was reported to provide similar savings in energy consumption (Chen et al., 2022; Cui et al., 2020; Tututi-Avila et al., 2017; Yang et al., 2019a). In comparison to the normal configuration, the SS configuration contains two additional degrees of freedom, i.e., the side-draw flowrate and side-draw location. The SS configuration also has the advantage of better control performance since it does not lose any control degree of freedom as in the cases of TCD and DWC, due to the combination of the two reboilers or condensers as one, which eventually deteriorates the control performance of the system. Today, there are already some industrial applications of SS distillation, which indicates the successful PI application at commercial level (Kiss, 2013; Li and Liu, 1999).

3. Current statuses PI applied to advanced distillation

In this section, we focus on the PI application to advanced distillation-based processes, particularly on ED, RD, and RED, which fall within the authors' research area of expertise. Generally, these processes (ED and RD) are far more industrially established in comparison to the other advanced distillation-based process such as membrane distillation (Gerbaud et al., 2019; Kiss, 2013; Ma et al., 2019; Sharma and Mahajani, 2002). For instance, Harmsen (2007) reported that there are more than 150 industrial RD processes operating globally today for different applications, such as the manufacturing of methyl acetate, cumene, ethylbenzene, fatty acid esters, polyesters, the hydrolysis of acetates, and the manufacturing of methylal. These are well summarized by Kiss et al. (2019). In contrast, membrane-assisted distillation has not been widely applied in industry due to their more complex design.

3.1. PI applied to ED

Both the conventional external and internal intensification techniques have been widely applied to the ED for azeotropic

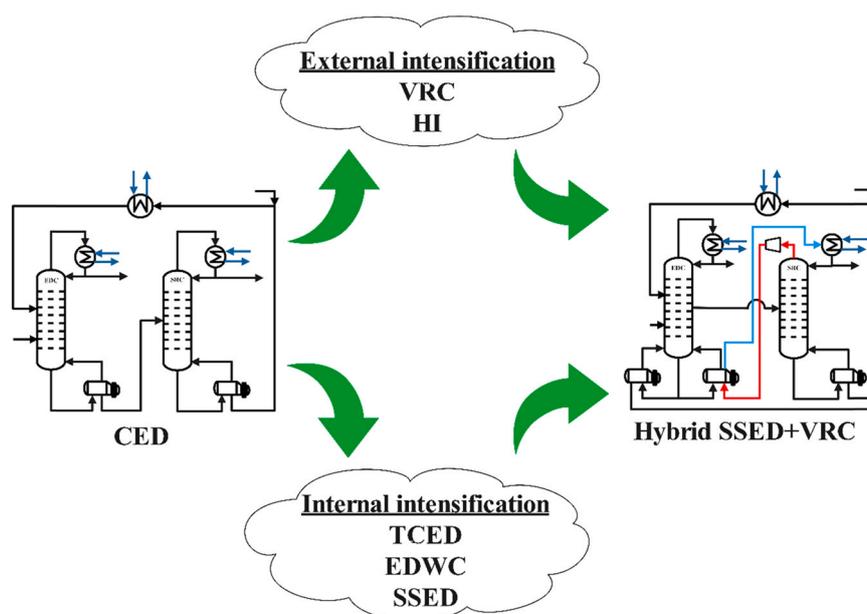


Fig. 2 – The evolution of PI techniques applied to ED.

separation (Wang et al., 2021a). Today, there are already several review papers that comprehensively summarize the application of PI to ED (Alcántara Avila et al., 2021; Kong et al., 2022a; Ma et al., 2019; Mahdi et al., 2015; Sun et al., 2019a) and therefore, this section aims to introduce only some of the novel intensified ED configurations recently developed. One novel configuration worth mentioning is the combination of SS with VRC as a dual intensification strategy to enhance the separation performance of the ED for the recovery of THF and ethyl acetate from waste effluent (Yang et al., 2022a). The dual intensification configuration was reported to provide significant enhancement in total annual cost (TAC) and CO₂ emissions of up to 33% and 49% with respect to the ED. For the sake of illustration, the evolution of such hybrid (i.e., dual) intensification from the CED is graphically exemplified in Fig. 2 while some other types of hybrid intensified ED processes are also available. Another example is the recent study that attempted to combine the thermally coupled with VRC for acetonitrile dehydration and reported that the dual intensified configuration enables significant savings in energy consumption and TAC of up to 30% and 20%, respectively (Cui et al., 2022). Another interesting intensification process was introduced by Jian et al. (2022) that integrates the preconcentration column (PC) with the solvent recovery column (SRC) for the recovery of ethyl acetate and isopropanol from wastewater effluent. HI or VRC were further applied to the novel intensified configuration as a heat recovery strategy, and they reported that the novel configuration coupled with VRC technique facilitates substantial improvement in energy and economic performance relative to the ED. Thus, it becomes clear that PI should not be limited to the application of a single internal or external intensification technique covered in earlier section to the existing ED process. Instead, PI can also involve combining several conventional intensification techniques together to become a novel dual intensification technology (e.g., the work of Yang et al. (2022a)). Moreover, PI does not necessarily involve only the conventional intensification techniques but can involve simple integration of two or more columns in the existing process as one (e.g., the work of Jian et al. (2022)).

3.2. PI applied to RD

Analogous to the PI application on ED, various conventional internal intensification techniques such as the SS (Thotla and Mahajani, 2009), DWC (Kiss et al., 2012; Kiss and Suszwalak, 2012), and TCD (Lee et al., 2012a) and external intensification such as VRC (Kumar et al., 2013) have been widely applied to the RD for azeotropic separation. These PI applications on RD have been systematically summarized in a few reviews (Kiss et al., 2019; Shu et al., 2022). In addition to these conventional PI applications, there are also studies that combined two or more conventional intensification techniques to the RD for improving the separation process performance. One example of such hybrid intensification study is the thermally coupled VRC RD reported by Liu et al. (2015) for the transesterification of methyl acetate and n-butanol to produce n-butyl acetate. Other hybrid intensification studies include but not limited to the dividing wall VRC (Jang et al., 2019; Shi et al., 2017) and the thermally coupled HI RD (Sun et al., 2019b). To our knowledge, the implementation of dual intensification on RD have been applied much earlier (e.g., 2015) while similar studies on ED only appeared in the 2020s. Today, studies on PI for RD are moving towards some other advanced hybrid intensification RD processes such as the double dividing-wall RD (Qin et al., 2022) or the hybrid RED for the separation of azeotropic mixture.

3.3. Reactive-extractive distillation (RED)

The RED is a recently emerging application, which has become increasingly popular in the past 2 years, particularly for the separation of azeotropic mixture (Fig. 3). The RED combines the beneficial features of both reaction and azeotropic separation as one, analogous to the traditional RD, except for the fact that the RED requires an additional solvent to facilitate the azeotropic separation. Such application, to our knowledge, was pioneered by the Shen's research group in 2020 where they introduced a triple column RED (TCRED) for the recovery of tetrahydrofuran and ethanol from wastewater (Su et al., 2020) and showed that the TCRED

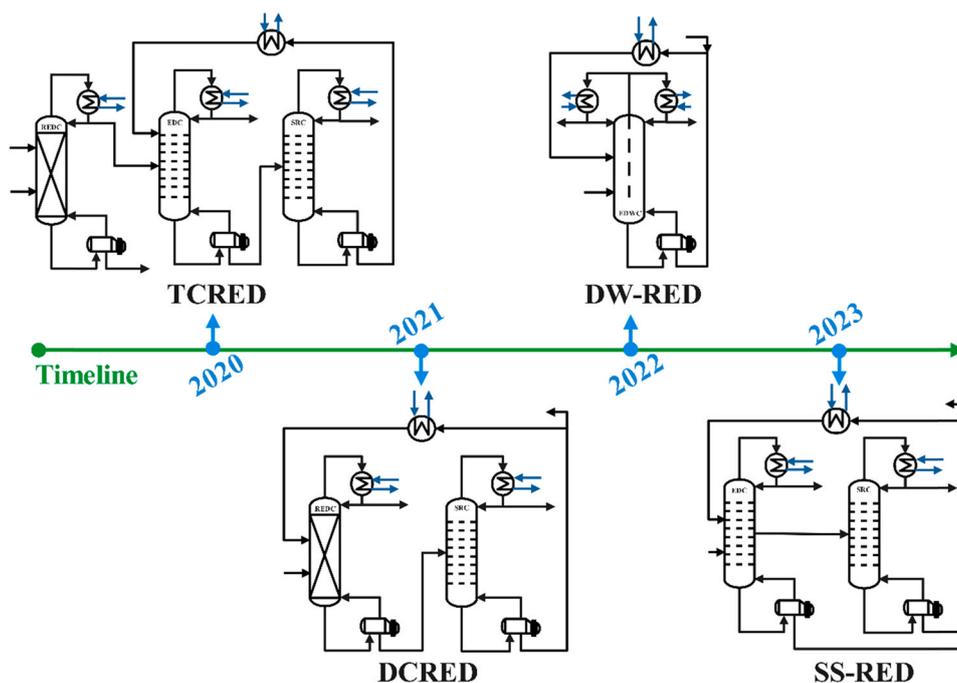


Fig. 3 – The evolution of RED (From left to right: Triple column reactive-extractive distillation (TCRED), Double column reactive-extractive distillation (DCRED), Dividing-wall reactive-extractive distillation (DW-RED), and Side-stream reactive-extractive distillation (SS-RED)).

provides significant reduction in TAC and CO₂ emission by 63% and 84%, respectively, relative to the original pressure swing distillation (PSD). In principle, two different RED configurations are available for the separation of ternary azeotropic mixture, i.e., the first configuration is a TCRED where the reactive and extractive separation processes take place sequentially in different columns while the second configuration is the double-column reactive-extractive distillation (DCRED) where both reactive and extractive distillation processes take place in the same column (Kong et al., 2022e, 2022d; Wang et al., 2021b). The detailed process flow diagram for these two configurations is given in our review paper (Kong et al., 2022c). Today, the RED has been widely applied to the separation and recovery of organic compounds such as ethanol and ethyl acetate (Wang et al., 2021b), tert-butanol (TBA) and ethanol (Zhang et al., 2021), and acetonitrile and isopropanol (Zhang et al., 2021) from wastewater. These existing studies have shown that significant improvement in TAC and CO₂ emission of up to 56% and 100%, respectively, can be realized. To further improve the performance of RED, traditional PI technologies such as TCD, SS, and DWC can also be incorporated to the RED. Shen's group incorporated the dividing wall to the RED, leading into a dividing wall reactive-extractive distillation (DW-RED) for the separation of TBA/ethanol/water (Yang et al., 2022b) and showed that there is a significant improvement in the capital cost. The same configuration (i.e., DW-RED) was later employed by other researchers for the separation of other ternary azeotropic mixture such as benzene/isopropanol/water (Yan et al., 2022) and ethyl acetate/ethanol/water (Liu et al., 2022). One notable drawback of the DW-RED however was the high operational cost because the DW-RED generally requires the use of high-pressure steam as heating utility, making it less economically attractive. To overcome this limitation, different energy conservation strategies such as feed pre-heating and ORC have been considered by existing study (Liu et al., 2022). Other than DW-RED, another alternative PI approach is the

use of a SS-RED, which has been demonstrated by Yang et al. (2023) to provide a lower TAC and CO₂ emission relative to the DW-RED for the recovery of isopropanol and ethyl acetate from wastewater.

3.4. Special cases

Until here, we have explicitly covered the application of different PI technologies to the RD, ED, and RED, and have shown how these PI technologies generate significant savings in the energy consumption and TAC. However, it is worth noting that there are also cases where the energy-intensified processes do not provide any energy-savings, which is counterintuitive to majority of the current studies. One notable example is the work of Yang et al. (2022b) that worked on the RED for the recovery of TBA and ethanol from waste effluent. Although they showed that the DW-RED enables significant improvement in TAC and CO₂ emission, the energy consumption was found to be slightly higher relative to the DCRED. Other than the work of Yang et al. (2022b), our literature survey has found two other studies that demonstrated the same results (Liu et al., 2022; Zhao et al., 2018). Altogether, these three interesting studies have demonstrated that PI do not always provide significant conservation in energy consumption. Nevertheless, these existing studies did not unambiguously clarify why energy-saving cannot be realized through PI. One possible reason for such contradictory results, in authors' opinion, lies in the distillation column configuration. Jiang and Agrawal (2019) reported that the number of basic and TCD configurations available generally increases combinatorially as the number of components in the feed increases. These configurations, while all carrying out the same separation task, can have very different capital and operating costs. Moreover, due to their structural differences, some configurations are easier to operate and control than others. These concerns naturally raise the following question: which distillation configuration

design(s) is/are more attractive to build and operate? For the past decades, several ideas and solutions have been proposed for designing energy efficient and cost-effective multicomponent distillation configurations. These approaches all fall into the category of PI. One recent study by the authors' research group attempted to preliminary analyzed energy-saving efficiency of the different advanced distillation-based processes covering ED and RED (Kong et al., 2022b). Three different case studies were developed and analyzed together with the three existing studies mentioned earlier (Liu et al., 2022; Yang et al., 2022b; Zhao et al., 2018) and it was preliminary deduced that the interconnecting flowrate or composition and high column internal vapor flowrate are two important factors that impact the energy-saving efficiency in intensified processes.

3.5. PI and its impact in issues beyond energy

Today, there is a pressing need to transition to more sustainable social and technical systems. Environmental problems, such as the loss of biodiversity, air, water, and soil pollution, the depletion of non-renewable resources and the excessive use of land for cultivation purposes, are putting the support systems in imminent and irreversible danger of life on earth. Economic challenges, unregulated markets, and flawed tax incentive structures lead to increasingly frequent financial and economic instabilities for individual companies, entire economies leading to ever more imminent financial collapse. In this sense, the intensification of processes can be seen as a framework with goals to achieve economic guarantor, inherent environmental safety, control indexes, among others for industrial processes (Sánchez-Ramírez et al., 2019). To date, the impact of PI on advanced distillation (i.e., RD, ED, and RED) have centered on energy, economic, and environmental aspects, with only a handful number of studies that explored the impact of PI on some other sustainability aspects such as control and safety (Liu et al., 2022; Yang et al., 2022b). According to Jiménez-González et al. (2012), one should consider incorporating "green metrics" when designing a process towards the broader target of circular economy. Further, modification in the topology, due to process intensification, for the same process can also modify circular economy indexes (Jiménez-González and Constable, 2014). Recent environmental, economics, safety, and control considerations, based on a sustainable process framework, have encouraged the chemical industry to focus on technologies based on PI to achieve those goals. The importance of considering sustainability issues early in the design of intensified process can help to differentiate between processes that are easy and processes that are difficult to operate. Thus, incorporating "green metrics" should be considered when designing an intensified process towards the goal of ensuring good control properties as part of the sustainability criteria. The aim of this principle is simple enough — to prevent waste and safety issues by identifying process excursions as they occur. Real time analysis and process control are necessary to carry out this action. By doing so, there may be sufficient time to modify process parameters such that the excursion may be reversed and there is no subsequent impact on safety and the final product quality.

Eco-Indicator 99 (EI99) is based on the life cycle assessment and the approach was proposed by Goedkoop and Priensma (2000). The metric has been applied successfully

in screening different intensified distillation alternatives with the lowest environmental impact, which indicates that the intensified configurations provide better environmental impact (Torres-Vinces et al., 2020; Vázquez-Castillo et al., 2019).

An inherent security metric is appropriate for evaluating a process in a framework of an intensified sustainable process under circular economy concept (Geissdoerfer et al., 2017a, 2017b). Process safety can be quantified by the individual risk (IR) index. The IR can be defined as the risk of injury or decrease to a person in the vicinity of a hazard (Freeman, 1990). Bravo-García et al. (2021), Sánchez-Ramírez et al. (2020), Segovia-Hernández et al. (2020) have shown that intensified systems give better results in inherent safety when compared to conventional systems.

The condition number has been established as the index to evaluate the control properties of the intensified configurations. The calculation of the condition number has been carried out through the singular value decomposition (SVD) of the relative gain matrix. The condition number gives valuable information about the theoretical control properties and represents the sensitivity of the system to input uncertainty. Large values must be avoided because they imply that the system is ill-conditioned. Alcocer-García et al. (2020), Santaella et al. (2017), and Cabrera-Ruiz et al. (2017) have used this index and found that in general the intensification of distillation systems improves dynamic performance.

In comparison to the advanced-based distillation process, the sustainability studies on ideal distillation and their corresponding intensified process are far more established. For instance, Sánchez-Ramírez et al. (2022) examined the sustainability benefits of using TCD and DWC for the purification of methyl-ethyl ketone, covering the economic, environmental, safety, and control aspects. The same indicators were also commonly employed by several existing studies such as the evaluation of the benefits of using TCD and Petyluk configurations for the purification of acetone (Amezquita-Ortiz et al., 2022), the application of TCD, DWC, and Petyluk to the RD for sustainable purification of lactic acid (González-Navarrete et al., 2022), and the use of TCD and DWC for the recovery valuable components from effluents discharged from the nylon industry (Bravo-García et al., 2021). These studies that examined how PI helps in enhancing the sustainability performance, reflect the importance of assessing the advanced distillation from a sustainable perspective, which in the authors' opinion, is still lacking in the existing studies.

4. Future perspective

4.1. Towards integrating different PI as one system

Upon analyzing the PI application on the existing studies, one noticeable trend was that most of the existing studies today are moving towards reducing the energy, environmental, and economic impacts, as part of the sustainable production initiative, by combining two or more traditional PI techniques as an innovative hybrid PI system (i.e., process). One obvious example is the hybrid side-stream with VRC as a dual intensification strategy proposed by Yang et al. (2022a) that provides significant enhancement in energy, economic, and environmental performance relative to using side-stream alone, as elucidated in Section 3.1. Today, most of the recent studies always provide better energy efficiency,

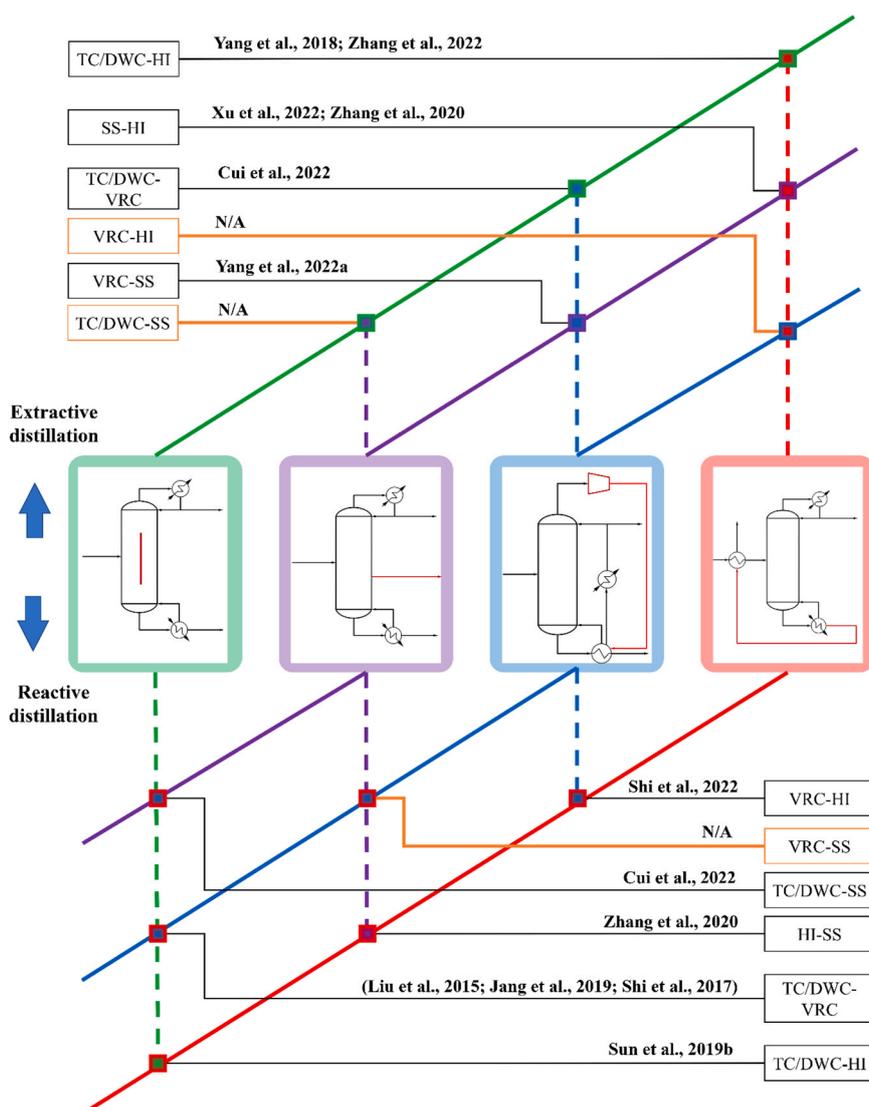


Fig. 4 – Summary of different PI combination applied to RD and ED.

economic, and environmental performance relative to the traditional (i.e., earlier) processes (e.g., the work performed in 2017 or 2018). Fig. 4 graphically summarizes the possible integration that combined the beneficial characteristics of two or more conventional PI technologies (e.g., SS, TCD, DWC) as one. Here, the TCD and DWC are classified under similar category since it has been well-reported that both TCD and DWC are thermodynamically equivalent to each other (Lee et al., 2019, 2012b). Based on our literature survey, existing studies have attempted to coupled TCD or DWC with VRC (Cui et al., 2022) or HI (Yang et al., 2018; Zhang et al., 2022) for ED, to further maximize the energy-savings. Likewise, there is also study on ED that combined the beneficial features of SS with VRC (Yang et al., 2022a) or HI (Xu et al., 2022) to maximize the energy recovery. To our knowledge, however, no existing study on ED have attempted to integrate the TCD with a SS to further enhance the separation process performance. Therefore, future study can explore the potential benefits of using a hybrid SS thermally coupled extractive distillation for the separation of azeotropic mixture.

For the intensification of RD system, it was observed from Fig. 4 that the dual intensification strategy is more widely established on RD than on ED. One important area of PI on RD worth further exploring is the application of SS because in authors' opinion, the application of side-stream to RD is less

studied. Our literature survey suggested that one of such study (i.e., SS RD) was conducted by Thotla and Mahajani (2009), which reported that the SS RD provides significant enhancement in product conversion and selectivity for the esterification of lactic acid and fatty acid. Another recent work on SS RD was reported by Yu et al. (2017) for the dimethyl adipate production and showed that the SS RD provides significant improvement in energy consumption and CO₂ emission. There are only three other studies, to our knowledge, that worked on the SS RD (Hasan et al., 2015, 2014; Nguyen and Lee, 2018). These studies signify the superiority SS RD as an energy-intensified process and thus, future research should be directed towards this area. Moreover, the control performance of the SS RD, to our knowledge, has never been investigated in previous studies. Other than SS RD, future research on PI for RD should also look into the mechanism of effectively integrating the reaction and separation, especially when RD is coupled with other intensification technologies, as suggested by Kiss et al. (2019). Shu et al. (2022) pointed out that although the modeling and simulation of both equilibrium and non-equilibrium stage models have been widely established for the RD process, the simulation results merely represent a micro-scale RD unit while the practical application of RD should be viewed from macro-scale perspective.

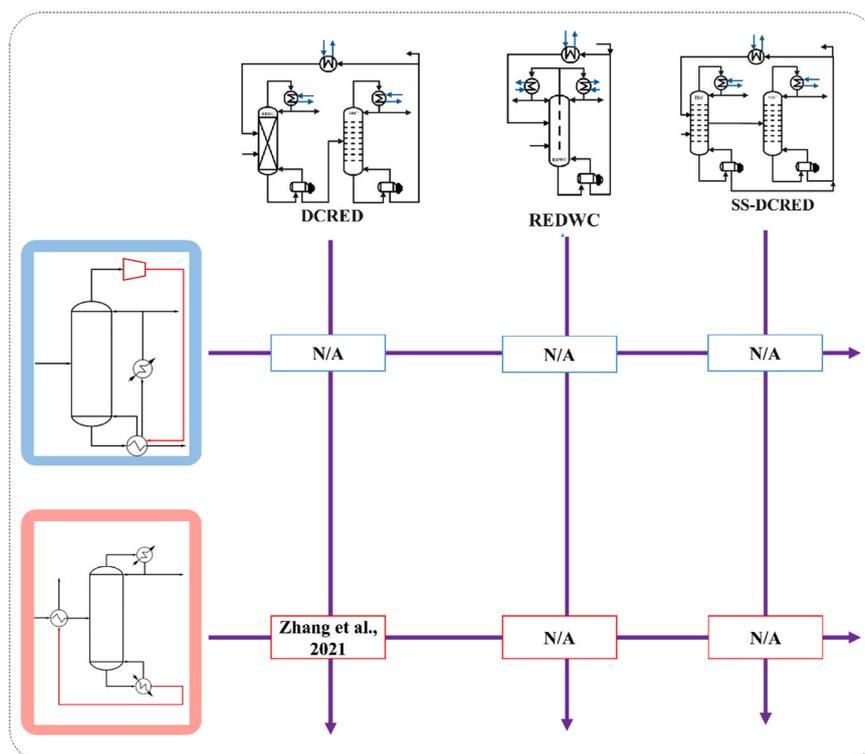


Fig. 5 – Summary of different PI combination applied to RED.

For the RED process, significant progress has been made towards applying different traditional PI techniques to increase the separation process performance (See Figs. 3 and 5). To date, the traditional PI techniques such as TCD, DWC, and SS have been applied to the RED, as described in Section 3.3. Nonetheless, no studies have attempted to further implement the VRC or HI as an external PI technique to the intensified RED (e.g., DW-RED or SS DCRED) for further maximizing the energy recovery, although the HI RED has been reported (Zhang et al., 2021). Therefore, future studies can consider maximizing the energy recovery through implementing VHRP or HI to the established energy-intensified RED.

4.2. Control performance of intensified RD, ED, and RED

From Section 3.1 and Section 3.2, it is apparent that many studies have already worked on the steady-state and dynamic studies for the RD and ED integrated with the widely established PI techniques such as TCD, DWC, and SS. Nonetheless, the control studies for the recently emerging novel intensified configurations such as those reported in the work of Jian et al. (2022) have not been explicitly studied. Although previous studies have demonstrated the superiority of these novel intensified configurations in providing a lower energy, economic, and environmental performance with respect to the conventional base case, it is not clear how robust and stable these novel configurations under operations and how the new system will respond under throughput or feed composition disturbance, which is important in the actual industrial operations. The operational controllability of these novel configurations is an important area worth further investigation because numerous previous studies have pointed out the existence of a trade-off between the steady-state (i.e., economic) and dynamic (i.e., control) performance for the RD and ED, even after these systems

have been intensified (Luyben, 2017, 2008; Wang et al., 2015). The best design that facilitates the best economic and environmental performance does not necessarily provide the best control performance (Alcántara Avila et al., 2021). To this end, it is imperative that future study must also consider the control performance as a part of the evaluation criteria whenever a novel configuration is developed. Alternatively, future study can also consider integrating both steady-state design and dynamic performance concurrently instead of using the conventional design sequence of “control after design” and these can be carried out using various techniques as covered by Alcántara Avila et al. (2021).

4.3. The future of RED

In previous section, we have elucidated the benefits of using RED as an energy-intensified separation process for ternary azeotropic mixture and demonstrated its energy, economical, and environmental superiority over the conventional RD or ED. The extended application of different conventional PI technologies such as TCD, DWC, and SS to the RED are also discussed. To our knowledge, however, the industrial application of RED for the ternary azeotropic mixture is still lacking, since the first simulation study of RED for azeotropic separation only surfaced in 2020 (Su et al., 2020). One key challenge worth highlighting is the presence of possible undesired side reactions in the complex RED system. Until here, all the existing studies on RED for ternary azeotropic mixture covered in previous section relied on the hydration of ethylene oxide (EO) to produce ethylene glycol (EG), where EG is generally used as solvent for subsequent azeotropic separation. During the EO hydration reaction, other side reactions may occur such as the reaction between EO and EG to form diethylene glycol (DEG), especially in the presence of excessive EO (Kong et al., 2022c). The situation can become even more sophisticated when RED is being implemented

into existing industrial process such as the ethyl acetate production. During such process, the presence of acetic acid in the EG or DEG can trigger other esterification reactions that form ethylene glycol monoacetate (EGMA) or even ethylene glycol diacetate (EGDA). All these side reactions can significantly affect the desired product purities and complicate process design. Another key barrier for industrializing the RED process also lies on the hydration reaction of EO itself. This is since EO is a highly flammable substance and thus, avoiding this highly flammable substance from escaping to the environment while maintaining the product standard is a challenging task. The industrialized RED therefore may require a stringent safety measure.

4.4. Energy efficiency of different energy-intensified (PI) configurations

In Section 3.4, we presented three interesting cases where PI do not always provide energy-saving, which contradicts most of the existing studies in literature. These three studies however did not provide any explanation on the inconsistency. In fact, our literature search has revealed that no studies to date can confirm under what scenarios the intensified processes present energy-savings and most importantly, determine the variables in the system that impact the energy-savings. Upon comparing these intensified systems for advanced distillation (i.e., ED, RD, and RED) against ideal (i.e., conventional) distillation system, it is important to point out that tremendous progress has been directed towards assessing the energy-efficiency of the intensified system for ideal distillation (Caballero and Grossmann, 2006, 2004; Giridhar and Agrawal, 2010a, 2010b; Gooty et al., 2019; Khalili-garakani et al., 2016; Mathew et al., 2019; Nallasivam et al., 2016). One notable example is the ease of separation index (ESI) introduced by Tedder and Rudd (1978) as heuristic to characterize the energy-saving efficiency of the different configurations for ternary distillation. Tedder and Rudd (1978) associated the energy performance of the different TCD to a few main variables such as the feed composition, the relative volatility, and the product purity specification. Other than that, some other heuristics have also been developed for examining the energy performance of intensified ideal distillation system, which are discussed in several literatures (Agrawal and Fidkowski, 1998; Chen and Agrawal, 2020; Segovia-Hernandez et al., 2021; Shah and Agrawal, 2011). These heuristics nonetheless are only applicable to ideal saturated liquids. To this end, we recommend future study to work towards analyzing the energy-saving efficiency in these advanced distillation systems. The crucial step is to identify the variables in the system that impact the energy consumption performance and under what scenarios the energy-savings do not exist (Fig. 6). One potential starting point is to extend the heuristic developed previously for ideal distillation, for advanced distillation system. Nonetheless, a thorough confirmation must be carried out to ensure that the previously developed heuristic can be extended to the separation to advanced distillation systems.

4.5. PI beyond energy

As indicated in Section 3.5, most of the existing studies on PI for advanced distillation (i.e., RD, ED, and RED) only prioritized the impact towards either energy, economic, and/or

environmental aspects, with only a few studies that considered the impact of PI on some other sustainability aspects such as control and safety (Liu et al., 2022; Yang et al., 2022b). Thus, future study should equally consider the impact of PI towards all sustainability indicators, that include economic, environmental, safety, and control metrics, which are all important features that a sustainable process must possess according to several review papers (Constable et al., 2002; Curzons et al., 2001; Jiménez-González et al., 2012) (Fig. 7).

4.6. PI, and its link with industry

Industry 4.0 (I4.0) is a concept established in 2011 to transform how diverse process systems interact inside an integrated framework. The implementation of smart or intelligent technological infrastructures, such as Internet of Things (IoT), cloud computing, advanced fabrication tools, mobile technology, Artificial Intelligence (AI), cybersecurity, big data and analytics, and Cyber-Physical Systems (CPS), has several advantages, including flexibility and customization, profitability, safety, optimization, and productivity (Sharma et al., 2021). Thus, a substantial overlap between I4.0 benefits and PI major goals is present.

As a result, the convergence of I4.0 (CC-I4.0) technologies that can use Circular Chemistry, connects the cutting-edge technology to systemic changes in the industrial, economic, and environmental framework. Data from the separate operations (important process parameters and variables) is often collected to observe, manage, and forecast the behavior of those activities. The models and simulations of the many phenomena functioning on the system must be contextualized, analyzed, and consequently understood (insights), which call for an information flow (with greater data quality) inside these processes.

In a RD system, for instance, several sensors might offer the necessary data on the equipment's gradients in concentration, temperature, and velocity. Different intensified systems have quickly embraced these concepts. For instance, Li et al. (2020) use of a cloud-based lab, automation, and AI not only sped up the detection of chirality in perovskites (70% reduction in experimental time), but also enabled cross-collaboration without regard to geographical boundaries to spur the discovery of new materials.

Given everything said above, it is necessary to reconsider the PI strategy in light of the advantages of implementing I4.0 technologies and processes. Researchers in the field of chemical engineering have significantly influenced Machine Learning (ML) techniques, such as auto-associative neural networks (Kramer, 1992) and the generic Deep Learning (DL) network method.

The promise of ML for novel uses such as catalyst design has inspired chemical engineers (Goldsmith et al., 2018). In addition, the combination of low-cost, high-performance computing and communication platforms, continued automation of activities that are globally interconnected, tightening environmental constraints, and business expectations for faster delivery of products and services to market open up a great field of opportunity for automation of the chemical industry according to the expectations of industry 4.0. Computing modeling employing Quantum Mechanical (QM) techniques, such as density functional theory, can speed up the screening of catalysts by enabling fast prototyping and identifying active sites and structure-activity relationships.

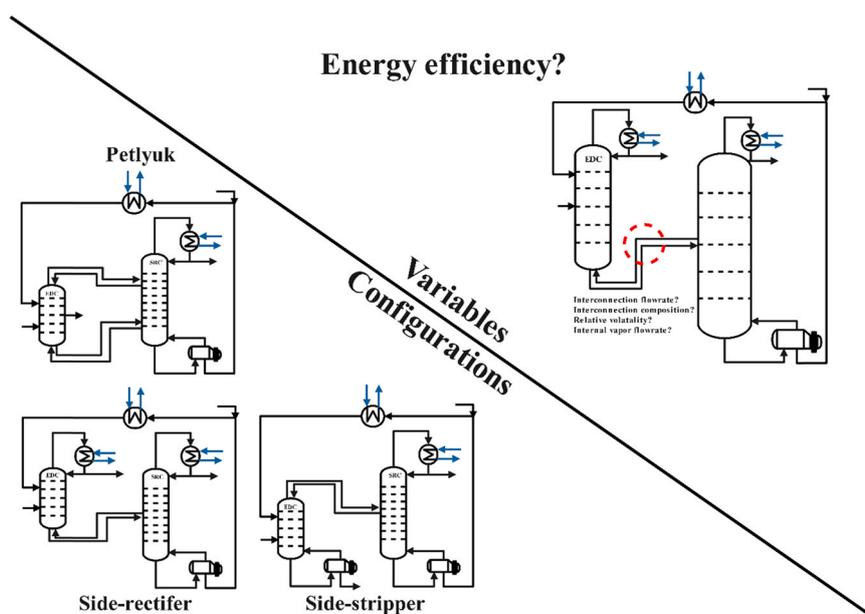


Fig. 6 – Illustration of the conditions that impact energy-savings in PI.

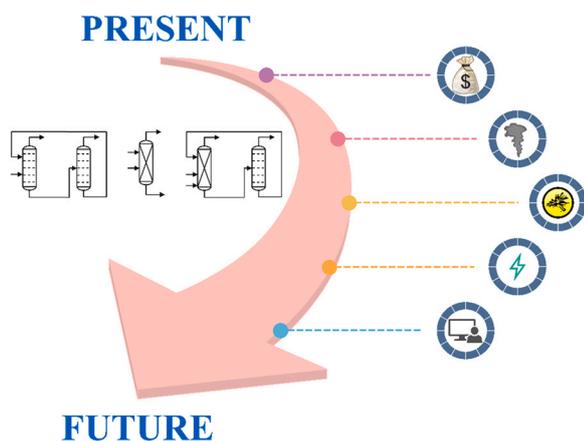


Fig. 7 – The future of PI beyond energy for ED, RD, and RED.

Additionally, an artificial neural network was used in the study by Sánchez-Ramírez et al. (2020) to simulate the dynamics of a wall column that divides an effluent from fermentation that produces acetone, butanol, and ethanol (ABE) for spark-ignition. A 4–10–3 multi-layer perceptron with a back-propagation algorithm proved sufficient to replicate the dynamics of the dividing column as reported by Aspen Plus Dynamics, considering the dynamics of the simulation. There were two different types of dynamic studies carried out: open-loop and closed-loop analyses. The expected vs actual values were bounded, according to the findings from the models that were validated. While most errors fell between 0.1% and 0.4%, the largest error percentage values were close to 0.9%. A one-layer neural network can depict the dynamics of an intensified column to predict the concentration of acetone, butanol, and ethanol, according to a study at later date by de la Fuente et al. (2022). Surprisingly, as activation functions, the linear activation function outperforms the tangent hyperbolic. In the end, we discovered that the reflux ratio and reboiler duty are essential components to reconstructing the complete dynamics of the intensified column.

5. Conclusion

This perspective paper focuses on the PI application for advanced distillation-based processes, featuring the ED, RD, and RED. Instead of discussing the technical aspects, we documented the contribution of PI applied to advanced distillation-based processes towards societal impact covering energy, economic, environmental, control, and safety perspectives. Starting with the historical application of PI to conventional distillation, we categorized the existing PI techniques available to date onto two categories, i.e., the first category are those techniques that require additional auxiliary equipment but does not involve altering the distillation configuration while the second category does. Other than general categorization, we also discussed the current statuses of PI for the advanced distillation-based processes, which are the main highlight of this work. In authors' opinion, one way forward for the application of PI to ED, RD, and RED, is to explore the beneficial features of integrating different traditional intensification techniques to become a hybrid intensification technology, instead of limiting the current application to a single internal or external intensification technique only. Other than prioritizing the economic and energy efficiencies as in the context of PI on traditional distillation processes, future study should be directed towards improving the sustainability performance of ED, RD, and RED, which are relatively less established relative to traditional distillation. Future work should also analyze the energy-saving performance in these advanced distillation systems by pointing out the variables that have direct impact on the energy-savings efficiency and under what scenarios the energy-savings do not manifest, which is another uncharted area for advanced distillation-based processes.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Authors gratefully acknowledged the support from Swinburne University of Technology Sarawak Campus and Universidad de Guanajuato. E. Sánchez-Ramírez and J.G. Segovia-Hernández appreciate the financial support provided by CONACYT (México).

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