CHEMICAL ENGINEERING TRANSACTIONS
VOL. 69, 2018

A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Elisabetta Brunazzi, Eva Sorensen
Copyright © 2018, AIDIC Servizi S.r.l.
ISBN 978-88-95608-66-2; **ISSN** 2283-9216

DOI: 10.3303/CET1869087

Hybrid Schemes for Intensified Chemical and Biochemical Process Alternatives

Nipun Garg[a], Anjan K. Tula[b,d], Mario R. Eden[b,d], Georgios M. Kontogeorgis[a*], John M. Woodley[a], Rafiqul Gani[c,d]

[a]Department of Chemical and Biochemical Engineering, Technical University of Denmark, DK-2800 Lyngby, Denmark
[b]Department of Chemical Engineering, 210 Ross Hall, Auburn University, Auburn, AL 36849, USA
[c]PSE for SPEED, Skyttemosen 6, DK-3450 Allerod, Denmark
[d]PSE for SPEED, 294/65 RK Office Park, Romklao Road, Bangkok, Thailand
gk@kt.dtu.dk

A practical way to generate innovative and sustainable designs of chemical and biochemical processes is to develop methods to generate hybrid schemes and apply them to solve specific energy intensive separation problems. Process intensification plays a major role within the process synthesis methods where the primary process objectives are to satisfy targeted process performance parameters including process economics and environmental impacts. The phenomena-based technique employing a multi scale method operating at unit operation (unit-op), functional/task and phenomena level is suited for achieving targeted process intensification. This paper presents a detailed systematic framework to determine new, innovative and more sustainable intensified flowsheet alternatives using combined phenomena and predefined intensified hybrid schemes consisting of conventional techniques such as distillation, reaction, membrane etc. In these hybrid schemes, the specified process objectives, which cannot be achieved by a single unit-op are achieved by combining multiple unit-ops all operating at their highest efficiencies to achieve the desired separation together with the targeted performance parameters. An overview of the key concepts and step-by-step workflow of the phenomena-based intensification method for hybrid separation schemes is presented along with its implementation and highlighting as well, new improved solutions of published case studies involving chemical and biochemical processes.

1. Introduction

The trend in process design in recent years has been the development and application of integrated processes combining reaction and separation operations in one or reduced number of units. This is motivated by the fact that it reduces the number of equipment and plant size, improves the process efficiency and hence better process economy. Hybrid schemes are a combination of either different separation and/or reaction operations constituting promising design options to carry out multiple tasks more efficiently than the individual operations. Distillation, for example, is one of the most used separation techniques in chemical process industry and being one of the most energy intensive unit operations, it has also been used in many hybrid schemes, such as, distillation-membrane (Stephan et al., 1995), divided wall column (Kiss et al., 2012) and reactive distillation (Buchaly et al., 2007). Tula et al. (2017a) recently proposed a general hybrid method based on distillation-membrane to identify the optimal hybrid scheme for any desired separation process incorporating distillation. Methods for process intensification, being analogously a synthesis-based approach, are classified into heuristic, mathematical programming and hybrid (Tula et al., 2017b). For the design and utilization of specific hybrid/intensified unit-op within a process, several methods have been proposed (Peschel et al., 2012; Siefert et al., 2012; Skiborowski et al., 2013). The design of classical/conventional unit operations is well understood and vastly supported by computational methods, but on the other hand, systematic generation of innovative and optimal process flowsheets including intensified/hybrid schemes is still a challenging task. This objective can be achieved by evaluating operations at the lowest level of aggregation i.e., phenomena level (Lutze et al., 2013), where underlying driving forces associated with tasks

Please cite this article as: Garg N., Tula A., Eden M., Kontogeorgis G., Woodley J., Gani R., 2018, Hybrid schemes for intensified chemical and biochemical process alternatives, Chemical Engineering Transactions, 69, 517-522 DOI: 10.3303/CET1869087

to be performed by the unit operations can be investigated to systematically generate alternative designs by incorporating and identifying intensified/hybrid schemes. As the synthesis problem is studied at a smaller scale, like atoms or groups of atoms combining to form many molecules, phenomena combining to form tasks that are combined to perform the desired operations, also create opportunities to generate new and innovative hybrid solutions for desired reaction-separation problem.

In this article, the phenomena-based synthesis approach (Babi et al., 2015) together with extensions from Garg et al. (2018) is further extended including predefined hybrid schemes for intensified process alternatives. The developed framework is also expanded in terms of phenomena database, search space of unit-ops, algorithms to generate hybrid schemes, combination rules to identify basic structures translating to hybrid schemes consisting of classical unit-ops. New and innovative hybrid schemes can also be generated using the feasibility and combination rules in this phenomena-based framework. The framework employs a multi-scale option where an existing process flowsheet is analysed and the phenomena representing the unit operations of the flowsheet are identified. The identified phenomena are added to a list of other phenomena that can also perform the same tasks in the process. They are then combined to form operations (existing, hybrid/intensified or new-innovative) that can perform the same tasks or combination of tasks performed in original process, thereby, providing innovative and more sustainable hybrid/intensified alternatives.

2. Overview of multi-level framework for phenomena-based process intensification

2.1 Concept of phenomena-based approach for hybrid schemes

A process flowsheet can be decomposed in three different scales (levels): unit operations scale (highest level of aggregation), task scale and phenomena scale (lowest level of aggregation). At the lowest level, most of the processes can be represented by 9 phenomena building blocks (PBBs): mixing (M), two phase mixing (2phM), reaction (R), cooling (C), heating (H), phase transition (PT), phase separation (PS), phase contact (PC) and dividing (D) (Lutze et al., 2013). Here, one or more phenomena can be combined according to a set of rules to fulfil the objectives of any task. For example, combining mixing (M), reaction (R) and cooling (C) phenomena leads to exothermic reaction [M(L)=R(L)=C], where L (liquid) represents the phase of reaction. These combined phenomena are called simultaneous phenomena building blocks (SPB's), which are further combined to generate basic structures translating to existing, hybrid or totally new unit operations. For example, as shown in figure 1 (a, b), considering SPBs of M=2phM=PC(VL)=PT(VL)=PS(VL); M=C=2phM=PC(VL)=PT(VL)=PS(VL) and M=H=2phM=PC(VL)=PT(VL)=PS(VL), with same basic structures translating to numerous conventional (distillation column) and hybrid (divided wall distillation column) schemes can be generated. In addition, two different basic structures can also be combined to generate further new hybrid schemes. For example, a basic structure containing SPB: M(L)=R(L)=C SPB can be combined with any combination of above SPB's to form new basic structure translating to reactive distillation (figure 1c) or reactive divided wall distillation.

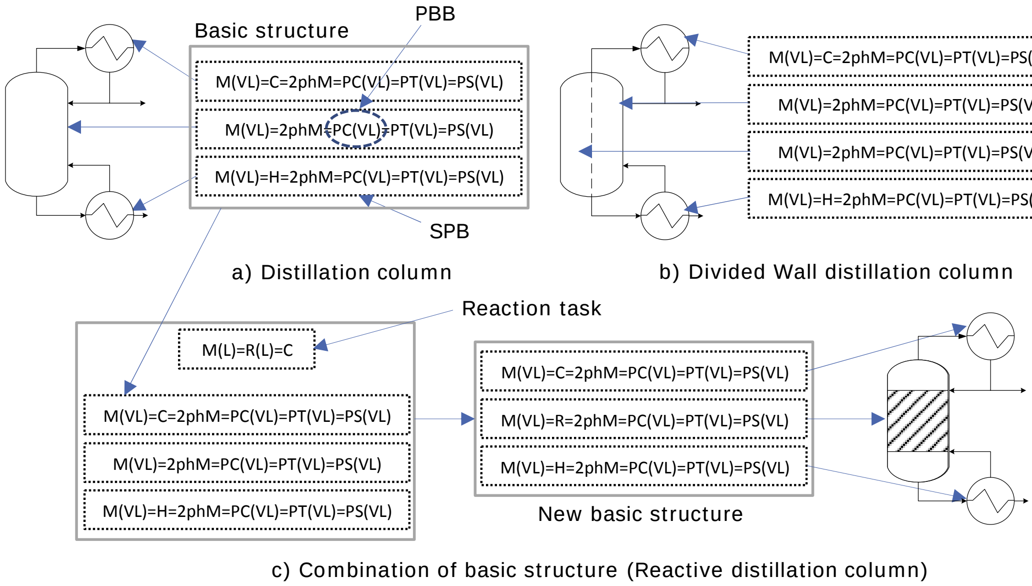

Figure 1: Illustration of concept behind phenomena-based synthesis for hybrid schemes

2.2 Workflow for phenomena-based synthesis intensification methodology

A systematic workflow diagram for phenomena-based synthesis intensification methodology is shown in figure 2 (adopted from Babi et al., 2015). The workflow consists of nine steps divided into 3 sections. The information flow, methods and tools used are also shown along with the steps.

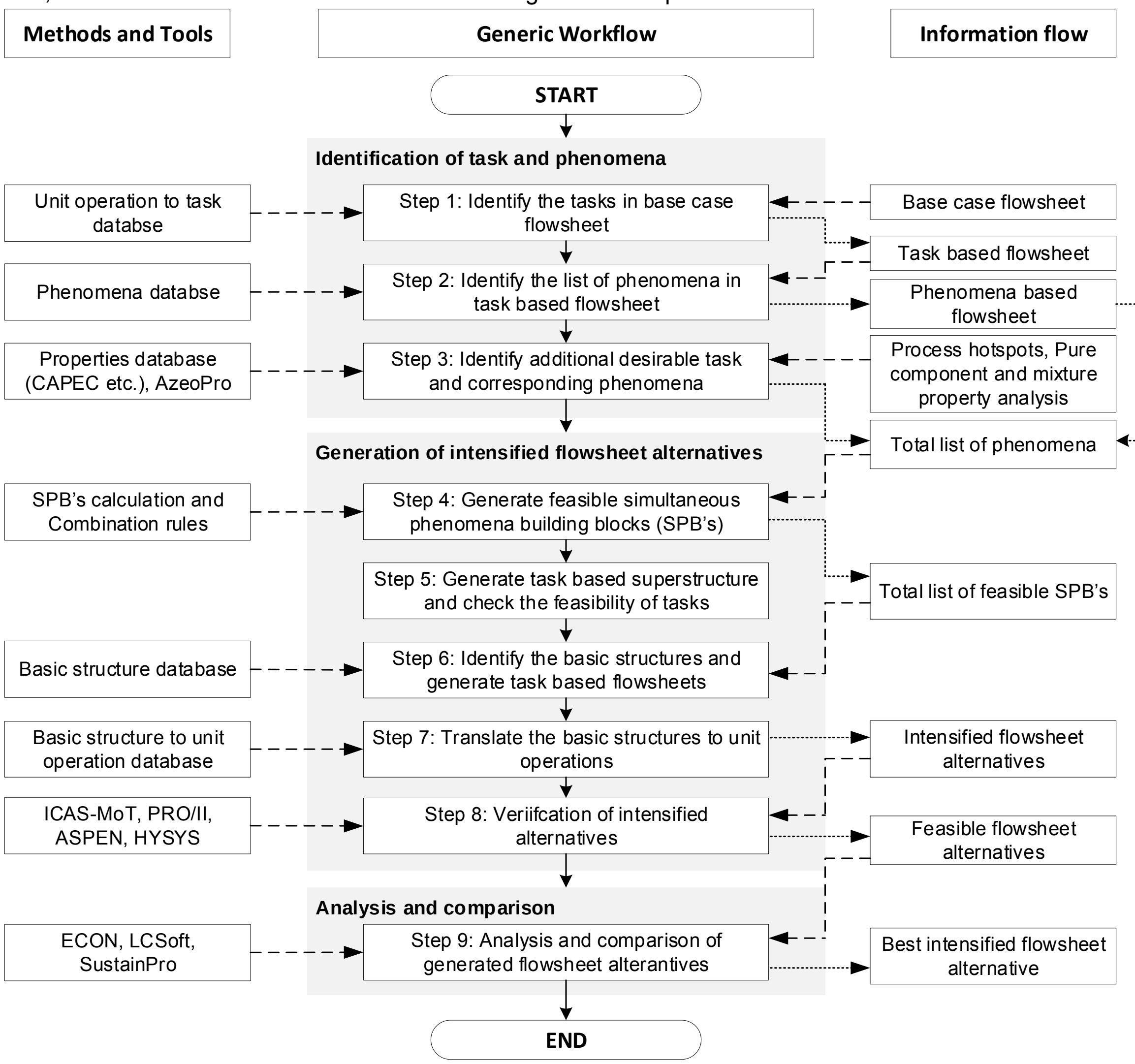

Figure 2: Workflow diagram for phenomena-based synthesis intensification methodology

Identification of desirable task and phenomena

In step 1, the base case flowsheet, which needs to be intensified, is translated to a task-based flowsheet. Here, each unit-op is assigned their corresponding task using database that translates unit-op to task. The unit-ops in the base case flowsheet are replaced by a single or a multiple task to obtain the task-based flowsheet. For example, the task performed by any reactor is reaction or by distillation, absorption etc. is separation. Then in step 2, corresponding PBB's involved in performing each task are identified using phenomena database to obtain the phenomena-based flowsheet. The total number of phenomena identified here is the initial search space, which is further expanded by identifying additional desirable task and phenomena. In step 3, the pure component and mixture property analysis data is retrieved from any property database to identify if there is/are any azeotrope or miscibility gap is present. After performing this analysis, additional desirable tasks and phenomena are identified using the database for identified process hotspots, tasks and PBB's linked to property ratio matrices of binary mixtures. Here, a desirable task is defined as a task that if performed has the potential to minimize or eliminate a process hot spot. These additional phenomena are then added to the existing list of phenomena, which then corresponds to the total list of phenomena with expanded search space.

Generation of intensified flowsheet alternatives

After identification of final list of phenomena, in step 4, total number of possible SPB combinations are calculated. However, not all combinations are feasible. Thus, feasible list of SPB's is identified using combination or connectivity rules. An example of a combination rule is that cooling (C) and heating (H) phenomena cannot be combined to form a SPB. Then in step 5, a task-based superstructure is generated to identify the minimum number of separation tasks that need to be performed and sequence the tasks starting from all possible reaction tasks to separation tasks. In step 6, the basic structures are identified that are able to perform a task, which are then listed as a feasible task with corresponding basic (SPB's) structures. Then using these sequence of tasks, feasible task-based flowsheets are generated. In step 7, these task-based flowsheets are translated into process flowsheets at the unit operation scale by using database to translate basic structures to tasks to unit-ops including predefined hybrid schemes. Here, if the basic structure and its corresponding unit-op do not exist, then in principle, a new unit operation is generated. Further, in step 8, a model-based analysis or detailed simulation analysis is performed to verify the feasibility of hybrid schemes for intensified alternatives.

Analysis and comparison

In the last step i.e., step 9, detailed economic, sustainability and life cycle analysis is performed in order to compare them with the base case and to identify the best intensified process alternative. Thus, alternatives that show improvements in all selected performance criteria (economic as well as environmental factors) as compared to the base case are non-tradeoff and more sustainable process flowsheet options.

3. Case study examples

The framework has been applied to solve many process synthesis problems, generating hybrid schemes for intensified chemical and biochemical processes (see table 1). Some of the recently reported case studies are listed in Table 1 together with brief overviews.

Table 1: Hybrid schemes identified for different chemical and biochemical processes using phenomena-based intensification

Process	Innovative alternatives	Hybrid/Intensified unit-op	Reference
Production of Isopropyl acetate	2	Plate-frame flow-pervaporator	Lutze, 2011
Separation of Hydrogen peroxide and water	1	Integrated distillation column	Lutze, 2011
Production of Cyclohexanol	1	Reactive distillation	Lutze, 2011
Production of Methyl acetate	4	Membrane reactor, reactive distillation	Babi et al., 2014
Production of Biodiesel	2	Membrane reactor, reactive distillation	Babi, 2014
Production of Dimethyl carbonate	4	Divided wall column, reactive distillation	Babi et al., 2015
Production of p-Xylene	4	Divided wall column	Anantasarn et al., 2016
Production of Ethylene glycol	1	Integrated membrane-distillation	Wisutwattana et al., 2017
Synthesis of Dioxolane products	3	Reactive distillation, reactive divided wall column	Castillo-Landero et al., 2017
Production of Bio succinic acid	3	Membrane bioreactor, membrane crystallizer	Garg et al., 2018

Production of Ethylene glycol (Wisutwattana et al., 2017) – In this case study, the base case design for the production of ethylene glycol through ethylene oxidation and ethylene oxide hydration reactions is available in the cited reference. The process mainly includes 2 reactors, 1 absorber and 2 distillation columns where 2 distillation columns are used to remove water and to purify ethylene glycol (98%) respectively. On performing detailed economic, environmental and life cycle analysis, it is analysed that, the highest utility cost (59% of total utility cost), equipment cost (42% of total equipment cost) and environmental impact (carbon footprint) is caused by one distillation column being used for water removal. Thus, by performing phenomena-based intensification, intensified solution involving hybrid schemes with membrane followed by distillation is identified (figure 3). As shown before, multiple basic structures generated from SPB's can perform same task. Thus, in this process, the separation task of water and ethylene glycol can be carried out by basic structures that consists of PT(VL), PT(VV) and PT(PVL) where PT(PVL) and PT(VV) translates to membrane. Recalculating the performance parameters using membrane, the utility consumption reduced by 45% while percent profit increased by 34% as compared to the base case that also led to 24% reduction in carbon emission.

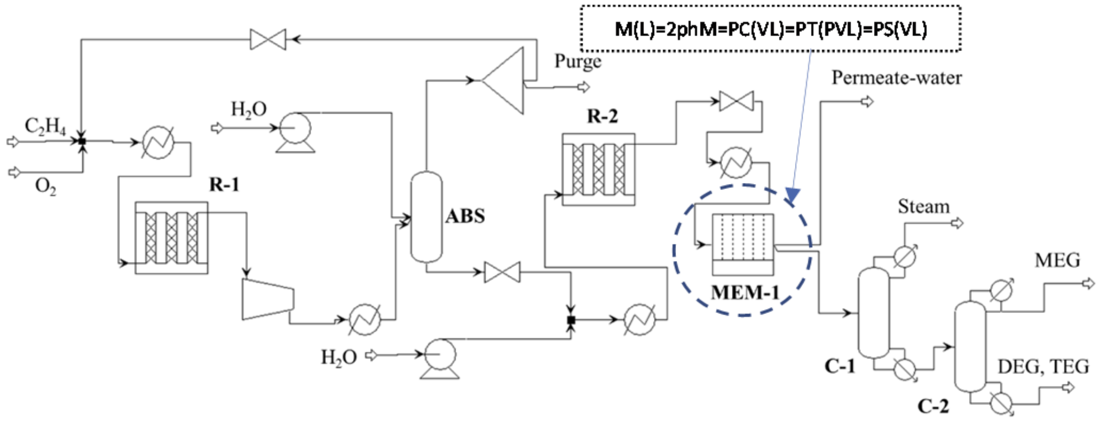

Figure 3: Intensified flowsheet alternative including membrane hybrid scheme for ethylene glycol production

Synthesis of Dioxolane products (Castillo-Landero et al., 2017) – In this case study, the base case flowsheet consists of an aldolization reactor followed by three distillation columns to get pure dioxolane products. The economic analysis and life cycle analysis showed that reboiler of second distillation column consumes 69% of total utility cost and has highest carbon footprint. Thus, using phenomena-based intensification, 3 intensified alternatives are generated. In alternative 1, the combination of reactor and first separation task was considered translating to reactive distillation. In alternative 2 last two separation tasks are combined into a single distillation column. In alternative 3, the combination of alternative 2 and 3 is considered to get a new basic structure translating to reactive divided wall column (RDWC) as shown in figure 4. In alternative 3, the overall utility cost reduced by 66% while carbon footprint reduced by 65%.

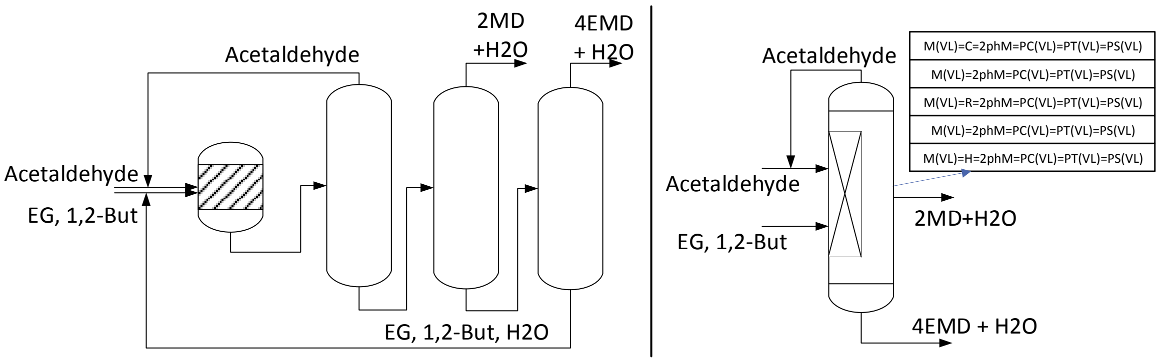

Figure 4: Base case vs hybrid scheme for best intensified alternative for synthesis of dioxolane products

Production of Bio-Succinic acid (Garg et al., 2018) – In this case study, the base case flowsheet is generated using a superstructure optimization approach followed by detailed simulation in PRO/II to get the base case design. The base case flowsheet includes single unit each of fermenter, centrifuge, distillation column, absorption column, crystallizer and dryer. Further, based on economic and life cycle analysis it is found that, the reboiler of the distillation column has the highest utility cost (73% of total utility cost) and has high carbon footprint. The sustainability analysis indicates loss of product and raw material in open path (in-out streams) containing crystallizer. These hotspots were further translated to design targets and thus by applying phenomena-based intensification, 3 intensified alternatives with different hybrid schemes are identified. In alternative 1, the combination of reaction task and immediate separation task i.e. removal of biomass is considered to obtain a new basic structure translating to hybrid scheme of membrane bioreactor. In alternative 2, the combination of last two separation task is considered leading to a basic structure translating to hybrid scheme of membrane crystallizer. Alternative 3 is a combination of alternative 1 and 2 (figure 5). Overall, recalculating the performance parameters for alternative 3, the utility cost reduced by 22% while global warming impact reduced by 23% thus a more sustainable solution as compared to the base case.

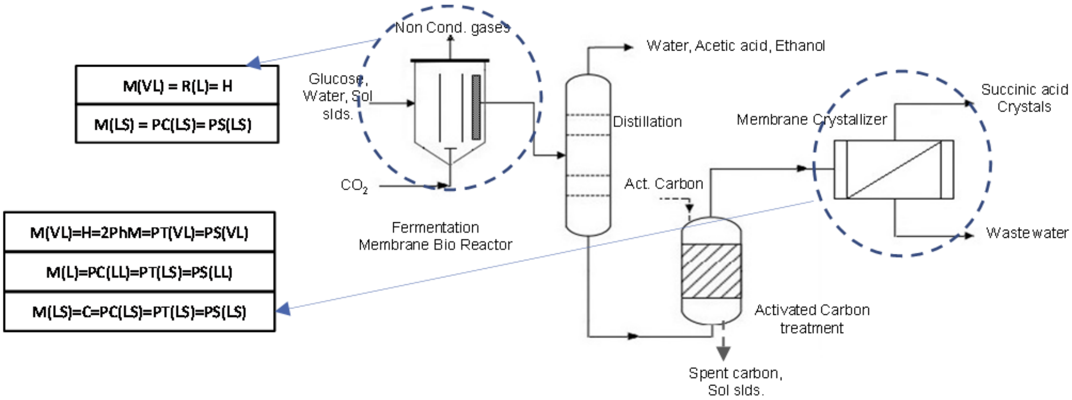

Figure 5: Hybrid scheme for best intensified alternative for production of bio succinic acid

4. Conclusions

A systematic phenomena-based methodology to generate hybrid schemes for intensified solutions has been presented. The framework includes extended search space of unit operations in terms of predefined hybrid schemes for intensified solutions. The application of the framework is illustrated through three case studies. It has been shown that by evaluating the problem at the lowest level of aggregation, new and innovative intensified solutions including hybrid schemes can be synthesized. In each case study, intensified solutions where either the downstream separation operations are combined or the reactor plus one or more downstream separation operations are replaced by predefined intensified hybrid schemes (reactive distillation, divided-wall distillation, crystallization-membrane, distillation-membrane, etc.). In each case, more than 20% savings at economic and environmental level are achieved as compared to base case and thus non-trade-off more sustainable and innovative solutions are found.

References

Anantasarn N., Babi D. K., Suriyapraphadilok U., Gani R., 2016. Computer Aided Chemical Egg. 38, 1093-1098.
Babi D. K., 2014, PhD Thesis, Technical University of Denmark, Lyngby, Denmark.
Babi D.K., Lutze P., Woodley J.M., Gani R., 2014. Chemical Eng. Process. Process Intensification 86, 173–195.
Babi D. K., Holtbruegge J., Lutze P., Gorak A., Woodley j. M., Gani R., 2015. Computers & Chemical Engg., 81, 218-244.
Buchaly C., Kreis P., & Górak A., 2007. Chemical Engg. & Proc.: Process Intensification, 46(9), 790-799.
Castillo-Landero A., Jiménez-Gutiérrez A., & Gani R., 2017. Computer Aided Chemical Engg., 40, 081-1086.
Garg N., Kontogeorgios G. M., Woodley J. M., Gani R., provisionally accepted at Process Systems Engg. Conference 2018, San Diego, USA.
Kiss A. A., & Ignat R. M., 2012. Separation and purification technology, 98, 290-297.
Lutze P., 2011, PhD Thesis, Technical University of Denmark, Lyngby, Denmark.
Lutze P., Babi D.K., Woodley J.M., Gani R., 2013. Ind. Eng. Chem. Res. 52 (22), 7127–7144.
Peschel A., Jörke A., Freund H., Sundmacher K., 2012. Computers & Chemical Engg. 31:150–154.
Seifert T., Sievers S., Bramsiepe C., Schembecker G., 2012. Chemical Engg. Proc 52:140–150.
Skiborowski M., Harwardt A., Marquardt W., 2013. Annual Revision Chemical Biomolecular Engg. 4(1), 45–68.
Stephan W., Noble R. D., Koval C. A., 1995. Journal of Membrane Science, 99(3), 259-272.
Tula A. K., Befort B., Garg N., Camarda K. V., Gani R., 2017a. Computers & Chemical Engg., 105, 96-104.
Tula A. K., Babi D. K., Bottlaender J., Eden M. R., Gani R., 2017b. Computers & Chemical Engg., 105, 74-95.
Wisutwattana A., Frauzem R., Suriyapraphadilok U., Gani R., 2017. Computer Aided Chemical Engg. 40, 1135-1140.