Simultaneous Synthesis of Processes with its Heat Exchanger Networks: P-graph Approach

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P-graph framework that was originally developed for processing network synthesis (PNS), has been extended to heat exchanger network synthesis (HENS). On the basis of the new formulation, PNS and HENS can be solved simultaneously by adapting the formerly developed P-graph algorithms. Therefore, the optimality of the overall synthesis procedure can be guaranteed, which is not possible for the traditional sequential procedures of PNS and HENS. The solution of a case study illustrates that the process structure determined by the simultaneous approach can be substantially different from the structure generated by the sequential approach, naturally, the former is the optimal solution.

1. Introduction

Heat exchanger network synthesis (HENS) has received significant attention over the last decades, thanks to its capability of decreasing the operating cost and environmental impact of processes. Heat integration was first introduced by Linnhoff and Hindmarsh (1983) who presented the Pinch Technology as a technique to determine the target for heat recovery. Afterwards, numerous contributions have been developed for HENS, either working further on the targeting approach or optimizing a super-structure based mathematical model. The available methods have been exhaustively reviewed in works such as elaborated by Gundersen and Naess (1988), which reviews the initial state of the art, and Klemeš and Kravanja (2013) who overview the advances of last decades in the topic. Moreover, the work of Furman and Sahinidis (2001) is a valuable contribution to understand the complexity of HENS.

Process Network Synthesis (PNS), is the task of determining the network of the operations in the process. PNS is primarily based on mathematical models and superstructure. It can be performed by mixed-integer linear or nonlinear programming methods, or by a combinatorial approach (Peters et al., 2003). The P-graph framework is a combinatorial tool for PNS that represents the superstructure (called as maximal structure) by a bipartite graph of materials and operating units. The framework is based on the fundamental combinatorial properties of feasible process structures. These properties are general and independent of the types of mathematical models of the operating units therefore, the framework is also general. P-graph framework has been selected for the integration of PNS and HENS for two main reasons; it is effective in synthesizing highly interconnected networks and it is capable of generating the n-best or all process networks in addition to the optimal one.

A PNS problem is specified by the sets of operating units with their mathematical models, the raw materials/utilities with their unit prices, and products with their required production. The objective function is also specified as a function of the parameters of the operating units and materials/utilities of the process. The combinatorial properties of the network of operating units of a feasible process are collected and formally described as set of five axioms (Friedler et al., 1992). Networks of operating units satisfying these axioms are called combinatorially feasible structures. Therefore, the network of each feasible process must be combinatorially feasible. Thus, the search for the optimal network can be reduced to the set of combinatorially feasible networks without the risk of losing the optimality (Peters et al., 2003), providing a big acceleration in

searching for the optimal process. There are several general algorithms of the P-graph framework. Algorithm MSG is for generating the maximal structure that corresponds to the commonly used superstructure. Algorithm SSG (Friedler et al., 1995) is for generating the whole set of combinatorially feasible networks, algorithm ABB is an accelerated branch-and-bound algorithm. These algorithms are considered as building blocks for the current work.

2. Simultaneous synthesis of a process and its heat exchanger network by a unified P-graph approach

The result of simultaneously solving the synthesis of a processing system and its heat exchanger network is usually superior to the sequential solution when the process is synthesized first then followed by the synthesis of its heat exchanger network. Naturally, the simultaneous approach can guarantee the optimality of the solution for the combined PNS and HENS problems. Therefore, the simultaneous solution is preferred. Both PNS and HENS are extremely complex, their combination is expected to be even more complex. Nagy et al. (2001) recognized that the combinatorial nature of the P-graph based PNS can conveniently be combined with a HENS method based on the compositions of temperature intervals. The simultaneous solution of PNS and HENS that was based on P-graph algorithms for PNS and a specific algorithm for HENS, proved to be useful. However, since the investment cost of a heat exchanger was considered to be proportional to the heat transfer area in Nagy et al. (2001), the cost function was not necessarily realistic. In the current work, there are two major improvements relative to the previous method. The cost function of a heat exchanger has been generalized to a fixed charge linear or nonlinear function, moreover, the HENS problem has been homogeneously embedded into the PNS problem resulting in a unified technique for generating the optimal solution of the simultaneously considered PNS and HENS problems. For illustration, let Figure 1(a) be the superstructure of a process synthesis problem showing the heating and cooling duties. Figure 1(b) shows the maximal structure as a P-graph, Tables 1 and 2 include the related data. Nodes O1, O2, and O3 of Figure 1(b) represent the operating units, while nodes C1, H1, and H2 represent the cooling and heating duties. The required amount of the product in the example is 50,000 t/y.
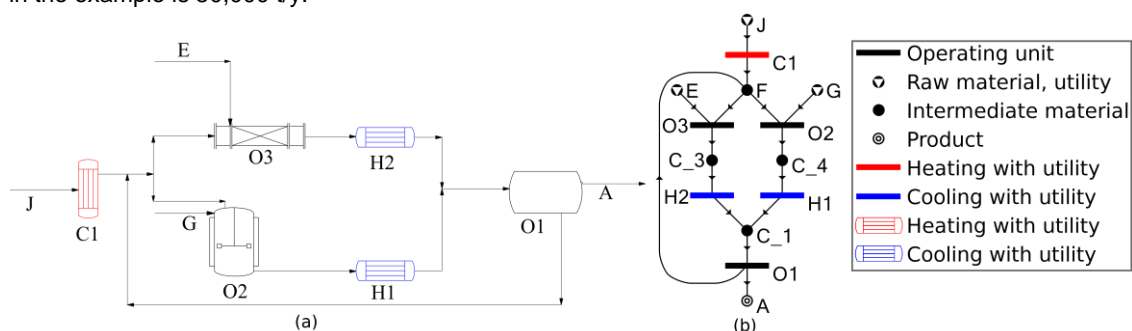

Figure 1: Illustrative example: (a) superstructure, (b) maximal structure represented as a P-graph

First, the hot and cold streams are split into substreams based on the initial and final temperatures of other streams as far as it has been described in Nagy et al. (2001). Then, composite streams are generated followed by extending the maximal structure with all possible heat exchangers including potential heat integrations (see Figure 2). The maximal structure with these heat exchangers is called integrated maximal structure. Since a material flow may have different temperatures troughout the realization of the process, the corresponding streams must be represented accordingly by different nodes on the integrated maximal structure (see e.g., H1_363 and H1_373). As an example, heat exchanger HE2 decreases the temperature of stream H1 from 373 K to 363 K, while increasing the temperature of stream C1 from 298 K to 353 K. The corresponding streams are denoted by H1_373, H1_363, C1_298, and C1_353, respectively.

Table 1: Heating and cooling duties of the illustrative example

Name	T_{in} (K)	T_{out} (K)	C_p (kJ/kg K)	h (kW/m^2 K)
C1	298	353	4.15	1.3
H1	373	308	2.37	1.3
H2	363	308	2.37	1.3

Table 2: Utilities of the illustrative example

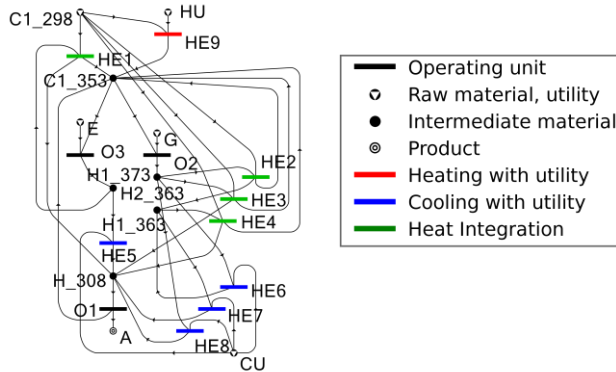| Name	T_{in} (K)	T_{out} (K)	Cost (USD/GJ)	h (kW/m² K)
Hot utility	373	373	11.88	1.7
Cold utility	293	308	0.77	1.7

Figure 2: Integrated maximal structure of the illustrative example: extended with all possible heat exchangers

The cost function of a heat exchanger is given in terms of the heat transfer area. The relation between the transferred heat and the area of the exchanger is determined by design equation Q=U*A*LMTD, where Q is the transferred heat, U is the overall heat transfer coefficient, A is the heat transfer area, and LMTD is the logarithmic mean temperature difference. The heat transfer coefficient U is the harmonic mean of the two individual film heat transfer coefficients denoted by h. Here we assume that the cost function is $10,000+800*A^{0.8}$, which is given by the Aspen's Energy Analyzer® software, with a pay-out period of 5 years.

Suppose that the integrated maximal structure is available. Also, the mathematical models of the operating units of the PNS part of the integrated maximal structure are given as before, i.e., the output streams of an operating unit are given as linear or nonlinear functions of the input streams and decision variables. For a heat exchanger of the integrated maximal structure, the mathematical model is given by the energy balance, i.e. transferred heat $Q=m*C_p*\Delta T$, for both of the related streams, where m is the mass flow of the stream entering the heat exchanger, C_p is the specific heat of the material of the stream, and ΔT is the change of the temperature of the stream. In the model, the decision variable for a heat exchanger is the transferred heat, i.e. Q. Since the algorithm generates a set of potential heat exchangers with both C_p and ΔT defined, the relation between the flow rate and the transferred heat is, consequently, linear. The cost function of heat exchanger is the sum of a constant (fixed charge) and a linear or nonlinear concave function of the heat transfer area.

After the operating units of the integrated maximal structure are given, algorithm ABB (Friedler et al., 1996) can directly determine the optimal network. If the mathematical models of the operating units of the integrated maximal structure are linear, the bounding procedure is LP, otherwise, NLP. The optimal network of the illustrative example is shown on Figure 3(a) as a P-graph, and on Figure 3(b) with the conventional representation.
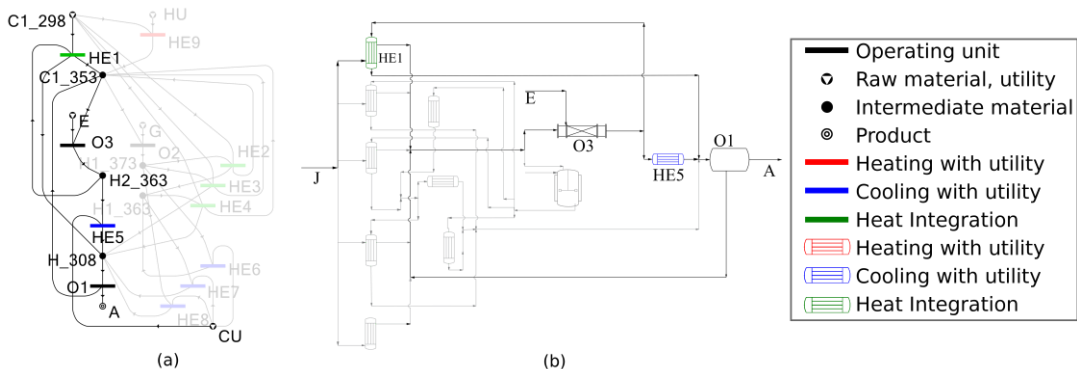

Figure 3: Optimal network of the illustrative example, (a) P-graph representation, (b) conventional representation

3. Case study

The simultaneous approach is employed to synthesize a methyl lactate process with a demand of 40,000 t/y. Methyl lactate is an ester usually present as an intermediate during the lactic acid's purification. It has gained attention as a bio-based solvent for lacquers and cosmetics (Aparicio, 2007). The maximal structure of Figure 4 covers all potential process networks for producing methyl lactate. The raw material streams, i.e., lactic acid and methanol are heated up to the reaction temperature and mixed with the recycled non-reacted components. The reactor's output is a mixture of methyl lactate, water, and non-reacted materials. Methyl lactate and non-reacted materials are recovered by distillations (units D1 through D8), which are considered as the feasible operations in the synthesis problem. Separation performs both product's purification and water removal. The unconsumed reagents are recycled and then mixed with raw materials. Because of the of lactic acid's side reactions, a purge of the side products is performed to prevent their accumulation in the process. Cost functions were obtained by simulating the operations in Aspen Plus®, and subsequently determining the investment and operating cost functions for each operating unit. The cost function of a heat exchanger is assumed to be $10,000+800*A^{0.8}$, as given by the Aspen's Energy Analyzer® software, and it is estimated here by the linear function $10,539+402.59*A$, with a pay-out period of 5 years.
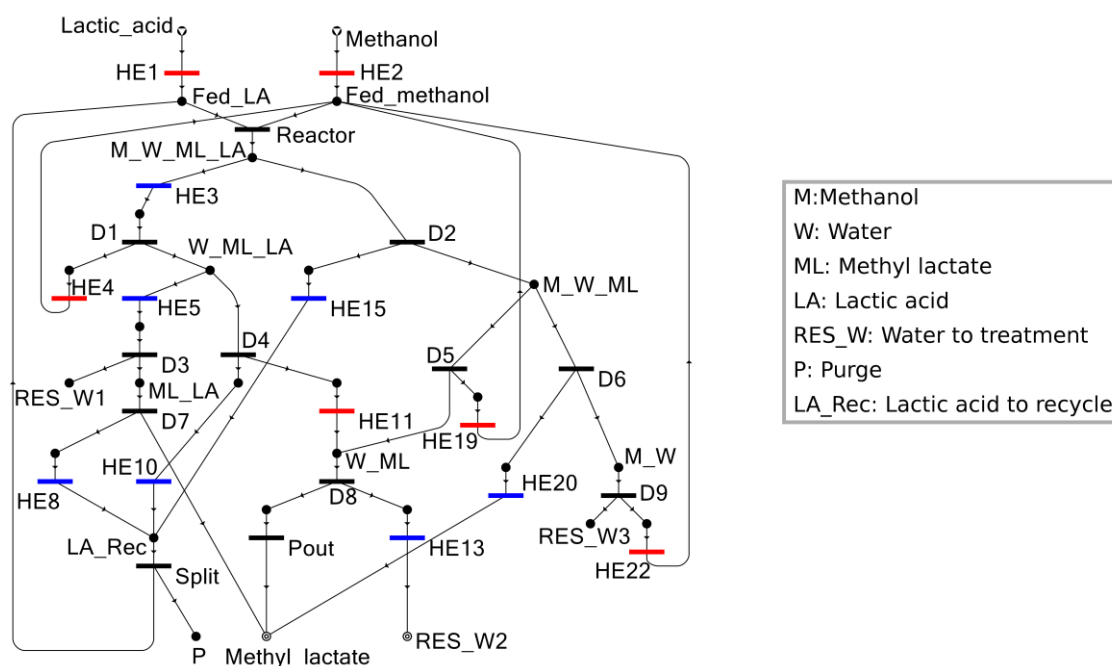

Figure 4: Maximal structure for the synthesis of methyl lactate process.

To reduce the cost of utilities for distillations, the temperatures of some streams are modified to be equivalent to that of their feed stage in the tower, thus, a significant number of streams (represented in Figure 4 with coloured nodes) constitute the potential sources or sinks of energy. All plausible sources and sinks of sensible heat were considered as input for the generation of the intervals of temperature and the composite sub-streams. The optimal solution of the simultaneous synthesis is given on Figure 5. For testing and verifying the result of the simultaneous synthesis, an exhaustive enumeration has been performed as a sequential approach on all feasible processing networks. To do so, PNS was carried out first via generating all feasible processes by algorithm SSG without considering heat integration, then, heat exchanger network was synthesized for each feasible network. There are 8 feasible processes for the case study, their minimal costs for both the non-integrated and the integrated process are given in Table 3. Note that for a non-integrated process all heating and cooling duties are performed by utilities, its minimal cost includes the costs of utilities and the related heat exchangers. In an integrated process of Table 3, the optimal heat integration is considered. Therefore, integrated process that has the lowest non-integrated cost is the optimal solution of the sequential method. For the case study, the optimal solution of the sequential method is process #1 with 43,168,722 USD/y cost (see Figure 6). The optimal solution of the synthesis problem is process #2 with 43,132,226 USD/y cost. This process is the same as the solution of the simultaneous PNS-HENS synthesis (see Figure 5). It is interesting to see that the utility cost of process #1 is 19,389 USD/y and that of process #2 is 11,831 USD/y.
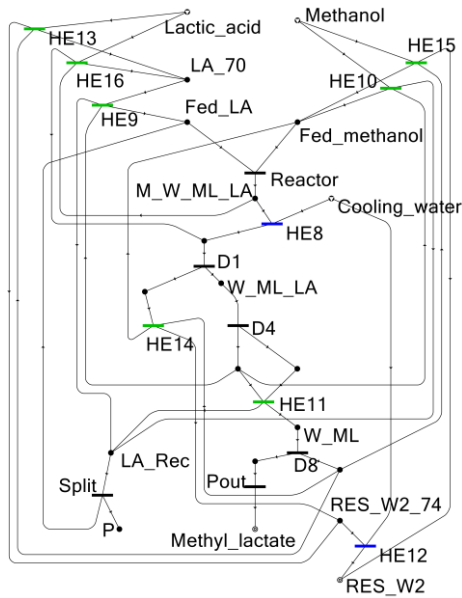
Figure 5: The optimal solution of the simultaneous approach (cost=43,132,226 USD/y)
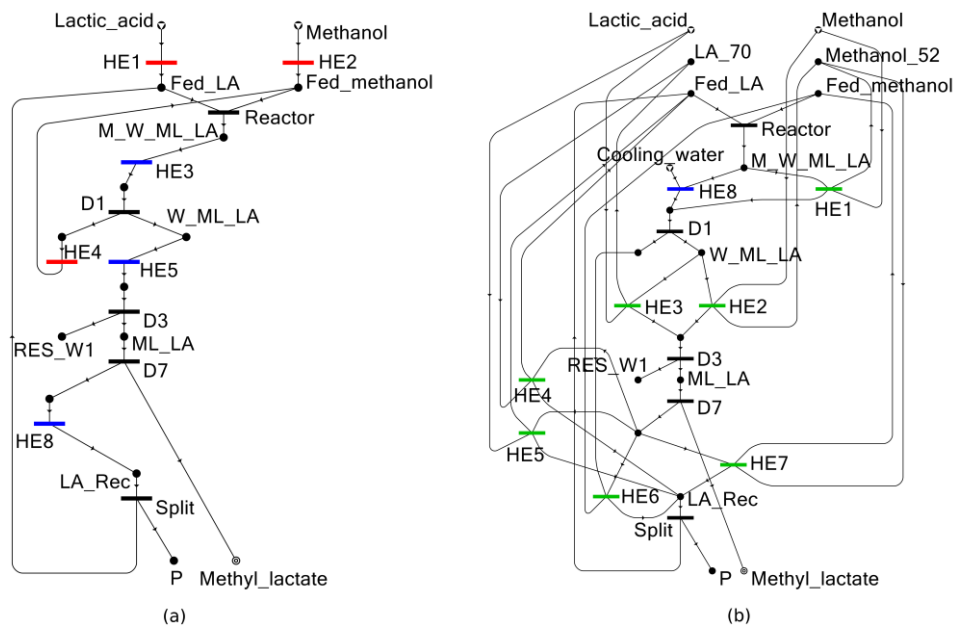

Figure 6: Process #1: the optimal solution of the sequential approach: (a) without heat integration (cost=43,400,700 USD/y), (b) with heat integration (cost=43,168,722 USD/y).

Table 3: Costs of the 8 feasible processes of the case study: without and with heat integration

Process #	Cost (Millions of USD/y)	
	Non-integrated	Integrated
1	43.400	43.168
2	43.450	43.132
3	43.990	43.751
4	44.432	44.194
5	46.316	46.021
6	46.974	46.686
7	46.977	46.635
8	47.753	47.479

The optimal process obtained with simultaneous approach differs from the one that would have been normally selected for the sequential approach (i.e. process #1), not only in the heat exchanger network, but also in the distillation sequence for the separation, which constitutes the 'core' of the process. In the process shown on Figure 6 (b), after recovering methanol in D1, the water is withdrawn by means of tower D3, and therefore, the product is obtained from D7. In the process of Figure 5, after methanol removal in D1, D4 separates lactic acid first, resulting in a lower temperature in the last stage of separation. Moreover, inclusion of heating requirement before D8 provides additional alternatives for heat integration. It is clear then, that including the feasible exchangers at the outset of the problem modified the synthesis problem, since the best processes obtained by the two approaches exhibit different separation sequences. This inference is only possible because of both approaches, i.e. the sequential approach with exhaustive enumeration, and the simultaneous approach, were performed, revealing an improvement of 36,450 USD/y when the simultaneous approach is employed. This demonstrates that under certain circumstances sequential approach is found to be insufficient to generate the best solution, hence joint synthesis of process and heat integration is necessary to guarantee the optimality of the process.

Some of the formerly developed algorithms of P-graph framework are freely available in P-Graph Studio, however, for solving the PNS and HENS simultaneously, the software for generating the integrated maximal structure is yet to be developed for P-Graph Studio. For this reason, the simultaneous synthesis has been performed here by a specific research software.

4. Conclusions

With the extension of the P-graph framework of processing network synthesis to simultaneous synthesis of processing networks and its heat exchanger networks, the systematic generation of the globally optimal solution of the combined problem has become available. The exhaustive analysis of a case study has shown that the sequential approach may generate considerably worse solution than that of the simultaneous approach. The capability of the proposed approach of generating the n-best processes in addition to the optimal one proved to be beneficial in practice.

Acknowledgement

References

Aparicio S., 2007, Computational Study on the Properties and Structure of Methyl Lactate, Journal of Physical Chemistry A ,111, 4671–4683.

Friedler F., Tarján K., Huang Y.W., Fan L.T., 1992, Graph-theoretic approach to process synthesis: axioms and theorems, Chemical Engineering Science, 47, 1973–1988.

Friedler F., Varga J.B., Fan L.T., 1995, Decision-mapping: a tool for consistent and complete decisions in process synthesis, Chemical Engineering Science, 50(11), 1755-1768.

Friedler F., Varga J.B., Feher E., Fan L.T., 1996, Combinatorially Accelerated Branch-and-Bound Method for Solving the MIP Model of Process Network Synthesis, In: State of the Art in Global Optimization, Springer, Boston, MA., 609–626.

Furman K.C., Sahinidis N.V., 2001, Computational complexity of heat exchanger network synthesis, Computers and Chemical Engineering, 25, 1371–1390.

Gundersen T., Naess L., 1988, The Synthesis of Cost Optimal Heat Exchanger Networks an Industrial Review of the State of the Art, Computers and Chemical Engineering, 12, 503–530.

Klemeš J.J., Kravanja Z., 2013, Forty years of Heat Integration: Pinch Analysis (PA) and Mathematical Programming (MP), Current Opinion in Chemical Engineering, 2, 461–474.

Linnhoff B., Hindmarsh E., 1983, The pinch design method for heat exchanger networks, Chemical Engineering Science, 38, 745–763,

Nagy A.B., Adonyi R., Halasz L., Friedler F., Fan L.T., 2001, Integrated synthesis of process and heat exchanger networks: algorithmic approach, Applied Thermal Engineering, 21(13-14), 1407-1427.

Peters M.S., Timmerhaus K.D., West R.E., 2003, Chapter 4, Plant design and economics for chemical engineers. McGraw-Hill, New York, USA.

P-Graph Studio <www.p-graph.org> accessed 16.05.2019.