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Software Tool for Formulating and Solving Various Process-Synthesis Problems

N. Sarkozi[1], B. Bertok[1], F. Friedler[1], and L. T. Fan[2]

[1]Department of Computer Science, University of Veszprem,
Veszprem, Egyetem u. 10, H-8200 Hungary
[2]Department of Chemical Engineering, Kansas State University,
Manhattan, KS 66506, U.S.A.

A language is introduced for formally defining process-synthesis problems. Moreover, a general framework is proposed for exhaustively and algorithmically generating all the candidate process structures for a problem without any assumptions except those explicitly stated in the problem definition. The generality of the method is illustrated by applying it to the synthesis of a reaction network and an azeotropic distillation system.

1. Introduction

The most essential question that must be answered at the outset of process synthesis, i.e., flowsheeting, is: which of the candidate, or plausible, connections among different operations should be taken into account, i.e., which of the outputs can be connected to which of the inputs? Most, if not all, available methods of process synthesis respond heuristically to this question. Even systematic or algorithmic methods assume the existence of an initial structure, or super-structure, containing all the candidate connections. This initial structure, more often than not, is constructed according to heuristic rules based on past experience. Exceptions are rare; see, e.g., Friedler *et al.* (1993) and Kovacs *et al.* (2000). In these works, the super-structure is generated algorithmically and is proved that a better solution of the problem cannot emerge by incorporating another structure.

Theoretically, an output from an operating unit implementing the desired operation can be connected to an input to another operating unit if there is a material that can be produced by one and consumed by the other. The materials can be specified by their properties, and the inputs and outputs are specified by the feasible ranges of the materials' property values. If the set of feasible property values of an output and the set of feasible property values of an input have a nonempty intersection, there

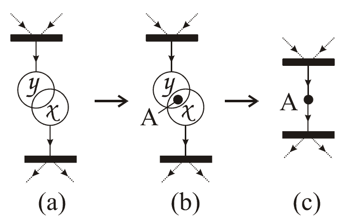


Figure 1. Connectivity of operating units.

exists a material with a certain property value that is feasible both as input and output; therefore, the output can be connected to the input. In Fig. 1, the operating units are represented by horizontal bars; and each of the sets of feasible property values of their inputs and outputs, by a circle. In Fig. 1(a), the set representing output y of one operating unit and the set representing input x of another operating unit have a nonempty intersection in the form of material. Thus, material A can be feasible as both output y and input x. In other words, output y can be connected to input x through material A. Material A is represented by a solid circle in Fig. 1(b) in the intersection of output y and input x. The resultant structure can be represented by a P-graph; see Fig. 1(c). P-graph is an unambiguous graphical representation of process structures (see, e.g., Friedler *et al*., 1992a, 1992b, 1993, 1995, 1996). Note that the properties of output y outside the intersection, e.g., material B, are further explored similarly for a possible connection to some other operating unit.

2. Problem formulation

Initially, the relevant properties of the streams, e.g., temperature, pressure, composition, and flow rates, are defined, thus resulting in a unique multidimensional representation of the materials. The outputs from and the inputs to the desired process system, i.e., the products and raw materials, and also the outputs from and the inputs to candidate operating units are given by the feasible ranges of their properties' values, e.g., a set of compositions and a temperature or pressure interval. This renders it possible to facilitate the systematic identification of various types of candidate operating units according to the regions of their feasible inputs and outputs and to automatically and rapidly examine whether an output from a unit of a certain type can be an input to another. This is a logical extension of the problem formulation given by Friedler *et al.* (1992a) for process synthesis where each of the feasible connections is explicitly given *a priori* in the problem definition by labeling the inputs and outputs among them by a single material.

The algorithmic method presented herein is illustrated by applying it to two synthesis problems different in nature, i.e., reaction-network synthesis (RNS) and synthesis of azeotropic distillation (AD) systems. The synthesis of vinyl chloride (see, e.g., Peters *et al.,* 2002) and the production of pure ethanol from its aqueous solution with toluene as the entrainer (see, e.g., Feng *et al.,* 2000) serve as the respective examples.

In RNS, any output from a reaction step (individual reaction) and any input to another can be connected if there is a material (reacting species), e.g., HCl in Fig. 2(a), which is an output (reaction product) from one and input (reactant) to another; see Fig. 2(b). A single node labeled HCl in Fig. 2(c) is the P-graph representation of the resultant connection. Singletons involving a single chemical species consumed and

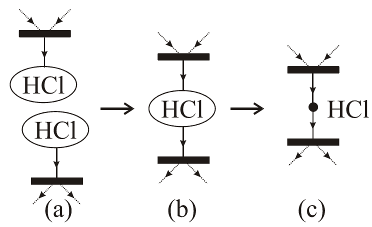


Figure 2. Connectivity of reaction steps in reaction-network synthesis (RNS).

produced define the inputs to and outputs from any reaction step. Each feasible connection can be explored algorithmically by searching for two sets: one representing an input and the other representing an output with a common intersection as described above. This entails searching for two singletons containing the same chemical species.

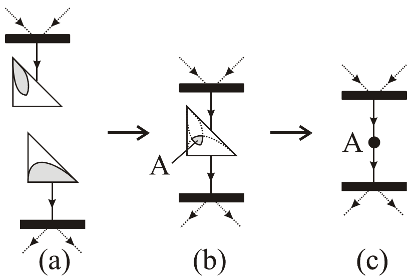


Figure 3. Connectivity of operating units in the synthesis of azeotropic-distillation (AD) systems.

In synthesizing an AD system, a feasible connection can be established between any output from an operating unit and any input to another if the intersection of the sets signifying the feasible ranges of the values of their properties is not empty. Figure 3(a) illustrates a gray field in the residue curve map (RCM) representing the feasible compositions of three components of an input and those of an output. These fields are overlapping, and thus, a set of compositions, e.g., material *A*, exists in the gray field in the RCM, each of which can be feasible as an output from one operating unit and an input to another; see Fig. 3(b). In other words, material *A* in this set can be produced by one operating unit and consumed by another. A single node labeled as *A* in Fig. 3(c) is the resultant feasible connection.

The synthesis problem is graphically defined in Fig. 4. Sets α's of inputs and sets β's of outputs of the operating units, desired products p_1, p_2, …, p_k, and available raw materials r_1, r_2, …, r_l are defined by the set of their feasible property values and represented by circles. Each operating unit is given by the set of its inputs and outputs; it is denoted by a horizontal bar. The problem is to synthesize a network of candidate operations leading from the raw materials to the desired products.

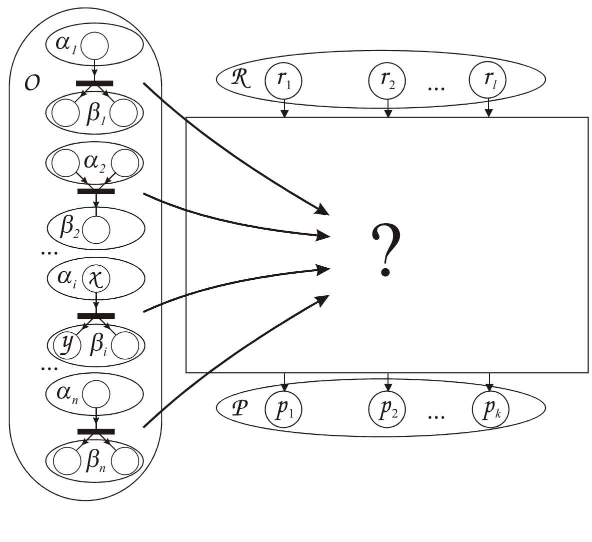


Figure 4. Definition of the process-synthesis problem.

In RNS, the inputs to and outputs from the reaction steps as well as the overall reaction are specified by singletons involving reacting species consumed and produced, i.e., the reactants and the products, respectively. In Fig. 5(a), the inputs to and outputs from a reaction step, especially the direct chloration, are represented by singletons $\{C_2H_4\}$, $\{Cl_2\}$, and $\{C_2H_4Cl_2\}$. Direct chloration is a candidate reaction step for synthesizing vinyl chloride. The overall reaction is defined by singletons $\{C_2H_4\}$, $\{Cl_2\}$, and $\{O_2\}$ representing its inputs (starting reactants) and singleton $\{C_2H_3Cl\}$ representing its output (final product); see Fig. 5(b).

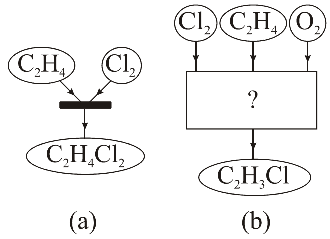


Figure 5. Definition of an RNS problem

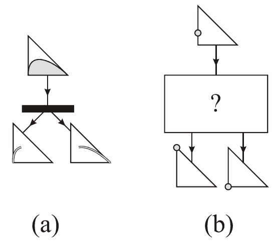


Figure 6. Definition of an AD-synthesis problem.

Feasible ranges of their compositions define the inputs to and outputs from candidate operating units for synthesizing AD systems. In Fig. 6(a), a decanter is depicted by the set of feasible compositions of its input and outputs, i.e., a liquid-liquid equilibrium envelope and its boundaries on the RCM. The input (feed-stream) to and outputs (product-streams) from the desired overall process are given by their composition, e.g., the composition of a two-component mixture and its pure components on the RCMs in Fig. 6(b).

3. Synthesis procedure

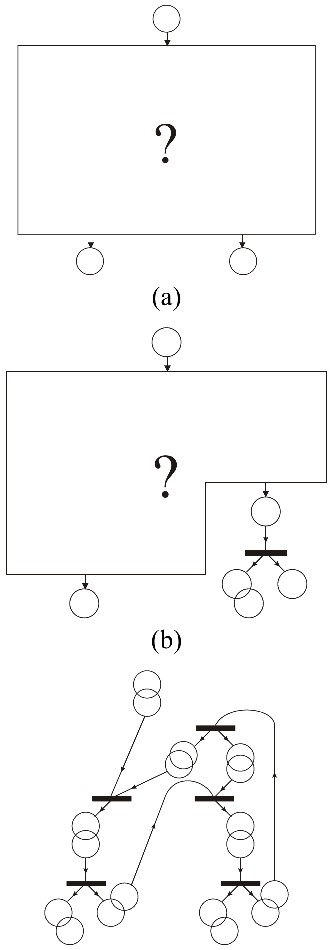
By resorting to the aforementioned language, an algorithmic synthesis method has been developed by merging the virtues of P-graph representation, algorithm SSG (Friedler *et al.*, 1992b) and the Means-ends analysis (Siirola, 1996). It generates feasible flowsheets in terms of combinatorial properties; mass balances; and constraints related to the inputs and outputs of the desired system and the operating units implementing certain operations. In contrast to algorithm SSG, feasible connections among the candidate operating units are explored during the execution: they are not explicitly given in the problem definition. The algorithm proceeds as follows:

The input to the algorithm is the synthesis problem as defined above. Initially, no process structure exists; therefore, the task is to generate networks of operating units leading from the raw materials to the desired products; see Fig. 7(a).

In the first step of the algorithm, an arbitrary final product is selected. An operating unit is incorporated into the structure if it is capable of producing this selected product, i.e., if the set of the feasible property values identifying one of its outputs intersects with the set of feasible property values representing the selected product. After the operating unit implementing a desired operation is included in the flowsheet, the set of required products is updated as necessary. Figure 7(b) shows that the selected final product is excluded from the set of required products while the input to the operating unit incorporated into the structure is included.

The generation continues on every possible branch enumerating all the alternative combinations of operating units capable of producing the materials of interest. The algorithm results in a feasible structure on a branch if the flowsheet represents a system where the desired products and inputs to the operating units in the structure are produced by consuming the given feed stream; see Fig. 7(c).

Finally, all the feasible flowsheets are generated algorithmically. They should satisfy the constraints related to the products, raw materials, feasible inputs and outputs of the operating units, and also those stated in the problem definition.



(a) (b) (c)

Figure 7. Steps of the synthesis method: (a) problem definition; (b) subproblem generation in the first step; and (c) resultant final structure.

4. Implementation

Process-network synthesis problems are formally defined by means of the Extensible Markup Language (XML). The language introduced is formally given by an XML scheme. The method for process synthesis proposed by resorting to the language introduced is implemented by means of computer programs. The well-defined language proposed enables algorithmic reformulation of various synthesis problems and serves as an open interface between these programs and other software packages. Such packages are those implementing algorithms MSG, SSG, and ABB (Friedler *et al.,* 1992b, 1993, 1995, 1996) for process-network synthesis or reaction-network synthesis; algorithm SNS-LMSG for separation-network synthesis (Kovacs *et al.*, 2000); the algorithm for generating feasible flowsheets for azeotropic-distillation systems (Bertok *et al.*, 2001); and algorithm PBT for reaction-pathway identification (Fan *et al.*, 2002). Note that executable codes implementing the algorithms mentioned above are downloadable from www.p-graph.com.

5. Application

The proposed method is applicable to a variety of process-synthesis problems, e.g., reaction-network synthesis, separation-network synthesis, and azeotropic-distillation-system synthesis. The proposed software has reproduced all flowsheets generated by specific purpose algorithms.

6. Conclusion

The proposed method gives rise to in-depth understanding of process-synthesis problems. The general-purpose computational tools are introduced in the current work in the form of software. They are applicable to various problems in the initial step of the synthesis instead of manually constructing mathematical-programming problems and prior to developing specialized solution algorithms implemented in programming languages. The set of candidate flowsheets generated by the general framework provides a benchmark comparable to those generated by a specific solution method.

7. References

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