Process Network Synthesis: Problem Definition

F. Friedler,[1] L. T. Fan,[2] B. Imreh[3]

[1] Department of Computer Science, University of Veszprém, Veszprém, Egyetem u. 10., H-8200, Hungary; Department of Chemical Engineering, Kansas State University, Manhattan, Kansas 66506

[2] Department of Chemical Engineering, Kansas State University, Manhattan, Kansas 66506

[3] Department of Computer Science, University of Szeged, Szeged, Hungary

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Abstract: Analyses of network problems have yielded mathematically and practically significant results. Naturally, it should be of substantial interest to extend such results to a general class of network problems where the structure of any system can be represented by a directed bipartite graph containing two types of vertices; the model for one of them is nonlinear. This class of problems is frequently encountered in the design of process systems for carrying out transformation of chemical or material species through physical, chemical, or biological means. General-purpose mathematical programming methods have failed so far to solve large-scale network problems involved in the design of such systems. This paper is intended to define this class of network problems, i.e., the problems of process network synthesis, and to elucidate the unique features of these problems. © *1998 John Wiley & Sons, Inc.* Networks **31:** 119–124, 1998

1. INTRODUCTION

In a process system, raw materials are consumed through various chemical, physical, and biological transformations to yield desired products; this is usually accompanied by the generation of wastes. Vessels in which these transformations are carried out are termed operating units of the process. A given set of operating units with the plausible interconnections can be described by a network. The desired products can often be manufactured using some subnetworks of this network. Thus, a given network may give rise to a variety of processes producing the desired products, and each of such processes corresponds to a subnetwork which can be considered to be its structure. Since the waste generation and energy and raw material consumption depend strongly on the selection of a process structure, the optimal design of such a process structure, i.e., the process network synthesis (PNS), has both economical and environmental implications. Studies of the network synthesis have given rise to significant results of a practical nature as summarized in the recent review article [4]. Nevertheless, these results are not directly applicable to PNS because of some of its unique features. The most significant among them is that the

Correspondence to: L. T. Fan; e-mail: fan@cheme.ksu.edu

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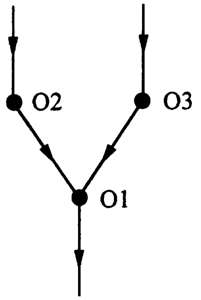

Fig. 1. Digraph representation of the structure of Examples 1 and 2.

transformation effected by an operating unit is frequently nonlinear; moreover, the optimal process structure may contain loops.

Since the mixed integer-nonlinear programming (MINLP) model of a PNS of a practical-size instance often contains an exceedingly large number of binary variables, the direct application of general mathematical programming methods is extremely difficult, if not impossible. Development of a method or methods exploiting the combinatorial features of PNS may be the only resource for circumventing this dilemma.

Our purpose is to present the general mathematical description and model of PNS. Since this model is often unsolvable for practical problems, it is reduced on the basis of the combinatorial properties of PNS.

2. A MINLP MODEL OF PNS

The simple directed graph is effective in representing structures of general network problems (see, e.g., [3]); however, it is unsuitable for PNS as demonstrated by the following examples (refer to Fig. 1).

Example 1. Two different intermediate materials are produced separately, one by operating unit o_2 and the other by operating unit o_3. Moreover, it is necessary to feed both intermediate materials into operating unit o_1 to generate the final product.

Example 2. One material is produced by both operating units o_2 and o_3. This intermediate is subsequently fed into operating unit o_1 to generate the final product.

Example 1 requires all three operating units to yield the final product, whereas, in Example 2, either a pair comprising operating units o_1 and o_2 or a pair comprising operating units o_1 and o_3 is sufficient to yield the final product. Nevertheless, the structures of these two examples are represented by an identical digraph as illustrated in Figure 1; the vertices correspond to the operating units and the arcs correspond to their interconnections. Similarly, representing materials by the vertices of a graph and representing their production and consumption by arcs do not define unambiguously the process structure. Structure representation with enhanced sophistication is required for PNS.

Process Graphs

Let M be a given set of objects, usually material species or materials that can be transformed into the process under consideration. Transformation between two subsets of M occurs in an operating unit. This operating unit is required to be linked to other operating units through the elements of these two subsets of M. The resultant structure can be described by a directed bipartite graph, termed a process graph or P-graph in short, which alleviates the difficulty encountered in representing a process structure by a conventional graph.

Definition 1. Let M be a finite set, and let set $O \subseteq \mathcal{P}(M) \times \mathcal{P}(M)$ with $M \cap O = \emptyset$, where $\mathcal{P}(M)$ denotes the power set of M. The pair (M, O) is called a *process graph* or *P-graph;* the set of vertices of this graph is $M \cup O$, and the set of arcs is $A = A_1 \cup A_2$ with $A_1 = \{(X, Y) | Y = (\alpha, \beta) \in O \text{ and } X \in \alpha\}$ and $A_2 = \{(Y, X) | Y = (\alpha, \beta) \in O \text{ and } X \in \beta\}$. P-graph (M', O') is defined to be a *subgraph* of (M, O), i.e., $(M', O') \subseteq (M, O)$, if $M' \subseteq M$ and $O' \subseteq O$. Let (M_1, O_1) and (M_2, O_2) be two subgraphs of (M, O). The *union* of (M_1, O_1) and (M_2, O_2) is defined by P-graph $(M_1 \cup M_2, O_1 \cup O_2)$ which is denoted by $(M_1, O_1) \cup (M_2, O_2)$. Obviously, this union is a subgraph of (M, O). If (α, β) is an element of O, then set α is called the *input-set* of (α, β), while

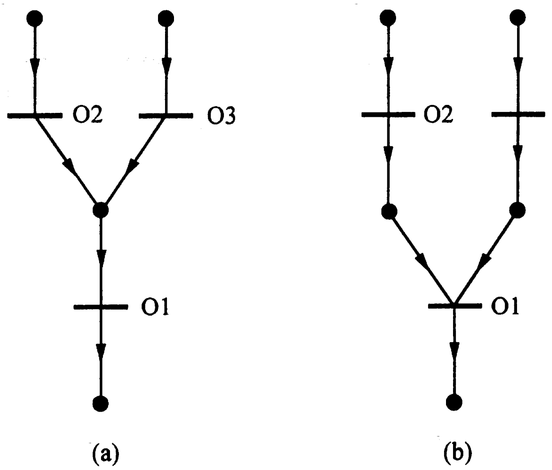

Fig. 2. P-graph representation of the process structure: (a) Example 1; (b) Example 2.

TABLE I. Plausible operating units of the practical example

No.	Type	Inputs	Outputs
1	F1	A1	A5
2	R1	A2, A3, A4	A9
	R1	A3, A4, A6, A11	A10
4	R1	A3, A4, A5	A12
5	R1	A3, A4, A5	A13
6	R1	A7, A8, A14	A16
	R1	A8, A14, A18	A16
	S1	A9, A11	A21, A22, A24
9	S1	A10, A11	A22, A24, A37
10	S1	A12	A25, A26
11	S1	A13	A25, A31
12	D2	A15, A16	A32
13	R1	A14, A17, A18, A19, A20	A33
14	R1	A6, A21	A35
15	W1	A22, A23	A48
16	W1	A5, A24	A36
17	S1	A5, A11, A25	A37, A38, A39
18	S1	A11, A26	A40, A42
19	R1	A14, A27, A28, A29, A30	A41
20	S1	A11, A31	A40, A42
21	C1	A32	A44, A45
22	W1	A33, A34	A46
23	S1	A36	A14, A48
24	S1	A38	A14, A48
25	F2	A41	A50, A51
26	W1	A43, A44	A53
27	F2	A46	A55, A56
28	S1	A47, A48	A5, A57
29	S1	A48, A49	A5, A58
30	S1	A50	A59, A60
31	D3	A51, A54	A61
32	D3	A52, A53	A61
33	D3	A54, A55	A61
34	D1	A59	A62, A63
35	S1	A60	A64, A65

set β is called its *output-set.* The sets of arcs incident into, out of, and to vertex X are denoted by $\omega^-(X)$, $\omega^+(X)$, and $\omega(X)$, respectively. The indegree, d^-, and the outdegree, d^+, of vertex X are defined by $d^-(X) = |\omega^-(X)|$ and $d^+(X) = |\omega^+(X)|$. The degree of vertex X is defined by $d(X) = d^-(X) + d^+(X)$. Since sets $\omega^-(X)$ and $\omega^+(X)$ do not intersect for a P-graph, $d(X) = |\omega(X)|$.

Figure 2 shows the two different P-graphs for the two examples that have identical digraph representation.

Preparation of the Model

Let us consider a process design problem in which the set of desired products is denoted by $\mathbf{P}$; the set of raw materials, by $\mathbf{R}$; and the set of available operating units, by $\mathbf{O} = \{o_1, o_2, \ldots, o_n\}$. Moreover, let $\mathbf{M} = \{m_1, m_2, \ldots, m_l\}$ be the set of the materials belonging to these operating units, and assume that $\mathbf{P} \cap \mathbf{R} = \varnothing$, $\mathbf{P} \subseteq \mathbf{M}$, $\mathbf{R} \subseteq \mathbf{M}$, and $\mathbf{M} \cap \mathbf{O} = \varnothing$. Then, P-graph $(\mathbf{M}, \mathbf{O})$, termed the *network* of the problem, contains the interconnections among the units, $o_1, o_2, \ldots,$ and o_n. Furthermore, each feasible process, producing the given set $\mathbf{P}$ of products from the given set $\mathbf{R}$ of the raw materials using operating units from $\mathbf{O}$, corresponds to a subgraph of $(\mathbf{M}, \mathbf{O})$, i.e., the *structure* of the process under consideration. For any $1 \le j \le n$, let $y_j = 1$ if o_j is contained in this subgraph and $y_j = 0$ otherwise. Thus, this subgraph is determined by the vector $(y_1, y_2, \ldots, y_n)$.

Let us now investigate the constraints with respect to the vertices and arcs of the network. For this purpose, let $A = \{a_1, a_2, \ldots, a_r\}$ be the set of the arcs and assign to arc a_k the continuous variable x_k $(k = 1, 2, \ldots, r)$ representing the quantity of either the material consumed or the product produced. The function for which $\varphi(\{a_{i_1}, a_{i_2}, \ldots, a_{i_t}\}) = (x_{i_1}, x_{i_2}, \ldots, x_{i_t})$ holds for any subset $\{a_{i_1}, a_{i_2}, \ldots, a_{i_t}\}$ of A is denoted by φ. Finally, variable z_j is assigned to operating unit o_j $(j = 1, 2, \ldots, n)$ for identification.

Operating unit o_j is linked to the system through interconnections represented by its arcs contained in $\omega(o_j)$. Especially, $\omega^-(o_j)$ includes the incoming arcs to vertex o_j, and $\omega^+(o_j)$ includes the outgoing arcs from vertex o_j. It follows that besides depending on y_j and z_j the constraint and cost of o_j depend only on the variables belonging to these arcs, i.e., on $\varphi(\omega^-(o_j)) \cup \varphi(\omega^+(o_j))$. Consequently, the constraints on the cost of operating unit o_j can be expressed, respectively, by

$$g_j(y_j, \varphi(\omega^-(o_j)), \varphi(\omega^+(o_j)), z_j) \le 0, \quad j = 1, 2,$$

$$f_j(y_j, \varphi(\omega^-(o_j)), \varphi(\omega^+(o_j)), z_j), \quad j = 1, 2,$$

where for a fixed value of y_j both f_j and g_j are nonlinear, differentiable functions on the practically interesting domain for $j = 1, 2, \ldots, n$.

Similarly, the constraint on and cost function of vertex m_i can be given, respectively, as follows:

$$g_j'(\varphi(\omega^-(m_i)), \varphi(\omega^+(m_i))) \le 0, \quad j = 1, 2, \qquad l$$

and

$$f_j'(\varphi(\omega^-(m_i)), \varphi(\omega^+(m_i))), \quad j = 1, 2, \ldots, l.$$

In practice, g' and f' are usually linear; the former represents the material balances and specifications of the products, e.g., quantity and quality, and the latter, the cost of raw materials.

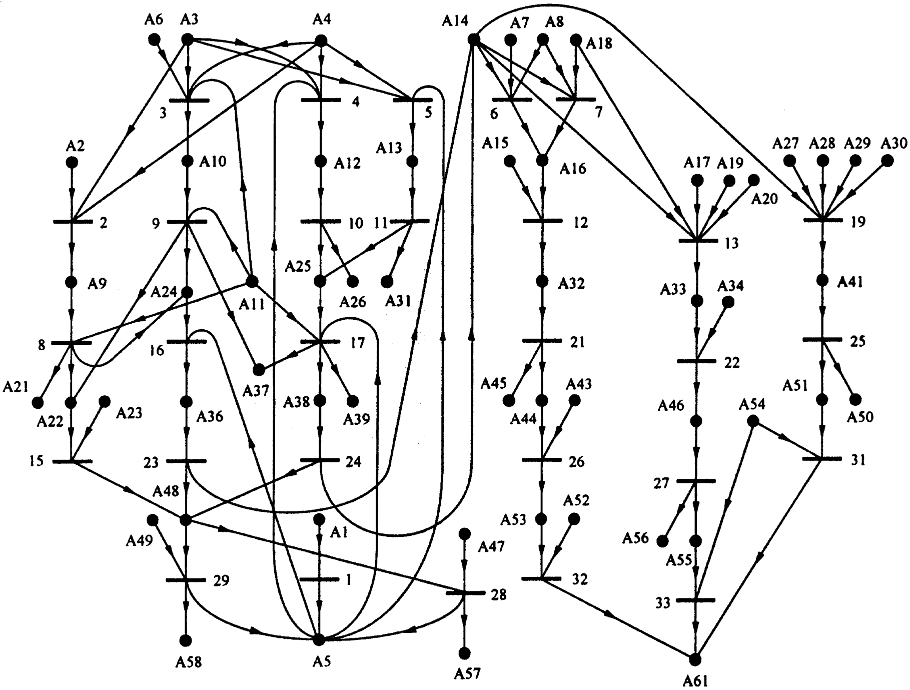

Fig. 3. Maximal structure of the practical example.

Process Network Synthesis Problem

Suppose that the nonempty finite set $\mathbf{M} = \{m_1, m_2, \ldots, m_l\}$ and triplet $(\mathbf{P}, \mathbf{R}, \mathbf{O})$ are given, where $\mathbf{P}$, $\mathbf{R}$, and $\mathbf{O}$ are nonempty finite sets and $\mathbf{O} = \{o_1, o_2, \ldots, o_n\}$. Also, suppose that $\mathbf{P} \cap \mathbf{R} = \emptyset$, $\mathbf{P} \subseteq \mathbf{M}$, $\mathbf{R} \subseteq \mathbf{M}$, $\mathbf{O} \subseteq \mathcal{P}(\mathbf{M}) \times \mathcal{P}(\mathbf{M})$, and $\mathbf{M} = \bigcup_{(\alpha,\beta)\in O} (\alpha \cup \beta)$. Then, the problem is to find

$$\left.\begin{array}{l} \sum_{j\in\{1,2,\ldots,n\}} f_j(y_j, \varphi(\omega^-(o_j)), \varphi(\omega^+(o_j)), z_j) \\ \quad + \sum_{i\in\{1,2,\ldots,l\}} f_j(\varphi(\omega^-(m_i)), \varphi(\omega^+(m_i)))\} \\ \text{subject to} \\ g_j(y_j, \varphi(\omega^-(o_j)), \varphi(\omega^+(o_j)), z_j) \le 0, \\ \qquad\qquad j = 1, 2, \ldots, n \\ g_i'(\varphi(\omega^-(m_i)), \varphi(\omega^+(m_i))) \le 0, \\ \qquad\qquad 1, 2, \ldots, l \\ y_j \in \{0, 1\}, z_j \ge 0, \\ \qquad\qquad j = 1, 2, \ldots, n \end{array}\right\} \quad (1)$$

3. A REDUCED MODEL OF PNS

One subgraph of $(\mathbf{M}, \mathbf{O})$ corresponds to each feasible solution of (1), the model of PNS. This subgraph is determined by vector $(y_1, y_2, \ldots, y_n)$, and it forms the structure of a process satisfying each constraint in (1), i.e., it is the structure of a feasible process or it is a *feasible process structure.* Obviously, not any vector $(y_1, y_2, \ldots, y_n)$, $(y_i \in \{0, 1\}, i = 1, 2, \ldots, n)$ defines a feasible process structure in general. However, the feasible process structures have some common combinatorial properties [1] that have been expressed implicitly in model (1). Since each feasible process structure must have these combinatorial properties, the set of subgraphs of $(\mathbf{M}, \mathbf{O})$, considered in solving model (1), can be reduced to the set of combinatorially feasible process structures or to the set of *solution-structures* in short.

Definition 2. Subgraph (M', O') of $(\mathbf{M}, \mathbf{O})$ is called a solution-structure of PNS if

(S1) $\mathbf{P} \subseteq \mathbf{M}'$, i.e., every final product is represented in graph (M', O');

(S2) $\forall x \in M'$, $d^-(x) = 0$ iff $x \in \mathbf{R}$, i.e., a vertex from M' has no input if and only if it represents a raw material;

(S3) $\forall u \in O'$, $\exists$ path $[u, v]$ in (M', O'), where $v \in \mathbf{P}$, i.e., every vertex from O' has at least one path leading to a vertex representing a final product; and

(S4) $\forall x \in M'$, $\exists(\alpha, \beta) \in O'$ such that $x \in (\alpha \cup \beta)$, i.e., any vertex from M' must be an input to or output from at least one vertex from O'.

The set of solution-structures is denoted by $S(\mathbf{P}, \mathbf{R}, \mathbf{O})$; its important properties are expressed by the following theorem, lemma, and corollaries.

Theorem. $S(\mathbf{P}, \mathbf{R}, \mathbf{O})$ is closed under union.

Proof. Let $\sigma_1 = (M_1, O_1) \in S(\mathbf{P}, \mathbf{R}, \mathbf{O})$, $\sigma_2 = (M_2, O_2) \in S(\mathbf{P}, \mathbf{R}, \mathbf{O})$, and $\sigma = \sigma_1 \cup \sigma_2 = (M', O')$. Hence, $M' = M_1 \cup M_2$ and $O' = O_1 \cup O_2$. It must be proved that conditions (S1)–(S4) are satisfied by σ:

(S1) $\mathbf{P} \subseteq M'$, since $\mathbf{P} \subseteq M_1$ and $\mathbf{P} \subseteq M_2$.

(S2) Let us define the following sets:

If $x \in M'$, $\bar{O} = \{(\alpha, \beta) | (\alpha, \beta) \in O' \text{ and } x \in \beta\}$,
if $x \in M_1$, $O'_1 = \{(\alpha, \beta) | (\alpha, \beta) \in O_1 \text{ and } x \in \beta\}$, and
if $x \in M_2$, $O'_2 = \{(\alpha, \beta) | (\alpha, \beta) \in O'_2 \text{ and } x \in \beta\}$.

(i) Suppose that $x \in M'$ and $x \in \mathbf{R}$.

(a) If $x \notin M_2$, then $\bar{O} = O'_1$; thus, $d^-_\sigma(x) = |\bar{O}| = |O'_1| = d^-_{\sigma 1}(x) = 0$.
(b) Similarly, if $x \notin M_1$, then $d^-_\sigma(x) = 0$.
(c) If $x \in M_1 \cap M_2$, then $\bar{O} = O'_1 \cup O'_2$; thus, $d^-_\sigma(x) = |\bar{O}| = |O'_1 \cup O'_2| \le |O'_1| + |O'_2| = 0$. From (a), (b), and (c), $d^-_\sigma(x) = 0$ if $x \in M'$ and $x \in \mathbf{R}$.

(ii) Conversely, let $x \in M'$ and $d^-_\sigma(x) = 0$. Then,

(a) If $x \in M_1$, then $\bar{O} \supseteq O'_1$; thus, $0 = d^-_\sigma(x) \ge d^-_{\sigma 1}(x) \Rightarrow d^-_{\sigma 1}(x) = 0 \Rightarrow x \in \mathbf{R}$.
(b) Similarly, if $x \in M_2$, then $x \in \mathbf{R}$.

From (a) and (b), we have $x \in \mathbf{R}$, if $x \in M'$ and $d^-_\sigma(x) = 0$.

(S3) $(M_1 \cup M_2, O_1 \cup O_2)$ contains all paths of (M_1, O_1) and (M_2, O_2).

(S4) This is implied by the next lemma. ■

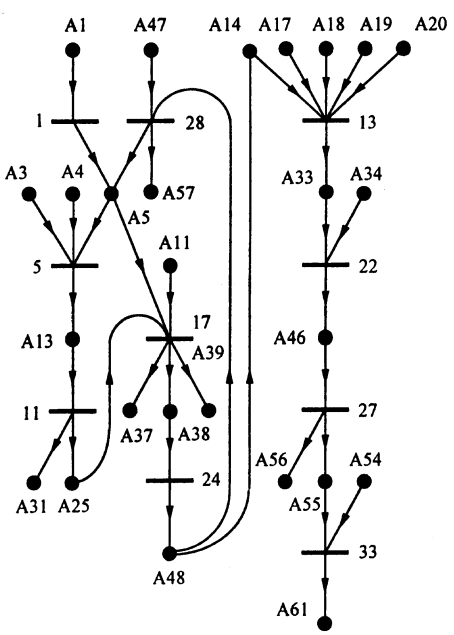

Fig. 4. Solution-structure for the practical example.

Lemma. If $(M', O') \in S(\mathbf{P}, \mathbf{R}, \mathbf{O})$, then $M' = \bigcup_{(\alpha,\beta)\in O'} (\alpha \cup \beta)$.

Proof. Since (M', O') is a P-graph, $O' \subseteq \wp(M') \times \wp(M')$ and

$$M' = \bigcup_{(\alpha,\beta)\in \wp(M')\times \wp(M')} (\alpha \cup \beta) \supseteq \bigcup_{(\alpha,\beta)\in O'} (\alpha, \beta)$$

Moreover, (S4) gives rise to

$$M' \subseteq \bigcup_{(\alpha,\beta)\in O'} (\alpha \cup \beta)$$ ■

The simple consequence of this lemma is the following corollary:

Corollary 1. Let $(M', O') \in S(\mathbf{P}, \mathbf{R}, \mathbf{O})$; then, (M', O') is uniquely determined if set O' is given. ■

The maximal structure of PNS, defined below, plays an essential role:

Definition 3. Let us assume that $S(\mathbf{P}, \mathbf{R}, \mathbf{O}) \neq \varnothing$; then, the union of all solution-structures of PNS, denoted by $\mu(\mathbf{P}, \mathbf{R}, \mathbf{O})$, is defined to be its *maximal structure,* i.e.,

$$\mu(\mathbf{P}, \mathbf{R}, \mathbf{O}) = \bigcup_{\sigma \in S(\mathbf{P},\mathbf{R},\mathbf{O})} \sigma.$$

Since the set of solution-structures is finite and closed under the union, the maximal structure also is a solution-structure; this leads to the following corollary:

Corollary 2. $\mu(\mathbf{P}, \mathbf{R}, \mathbf{O}) \in S(\mathbf{P}, \mathbf{R}, \mathbf{O})$.

Obviously, any operating unit not included in the maximal structure should not be considered for the optimal solution. Since any optimal solution is a solution-structure, the MINLP model of PNS can be based on the maximal structure. For this reason, let us suppose that $S(\mathbf{P}, \mathbf{R}, \mathbf{O}) \neq \varnothing$ and let the maximal structure, $\mu(\mathbf{P}, \mathbf{R}, \mathbf{O})$, be denoted by $(\hat{M}, \hat{O})$, and the set of the arcs of $(\hat{M}, \hat{O})$, by

$\hat{A}$. Furthermore, let the restriction of ω, ω^-, and ω^+ with respect to the set $\hat{M} \cup \hat{O}$ be denoted by $\hat{\omega}$, $\hat{\omega}^-$, and $\hat{\omega}^+$, respectively. Then, $\hat{\omega}(o_j) = \omega(o_j)$ and $\hat{\omega}(m_i) \subseteq \omega(m_i)$ are valid for any $m_i \in \hat{M}$ and $o_j \in \hat{O}$. In addition, let us constitute the following sets:

$$I = \{i: 1 \leq i \leq l \text{ and } m_i \in \hat{M}\},$$

$$J = \{j: 1 \leq j \leq n \text{ and } o_j \in \hat{M}\}.$$

Now, for any $i \in I$, let us derive the functions, F_i' and G_i', from f_i' and g_i', respectively, by assigning the value of zero to the variables belonging to the arcs from $\omega(m_i) \backslash \hat{\omega}(m_i)$. Then, we obtain the reduced model of PNS given below:

$$\left.\begin{array}{l}
\min\{\sum_{j \in J} f_j(y_j, \varphi(\omega^-(o_j)), \varphi(\omega^+(o_j)), z_j) \\
\qquad + \sum_{i \in I} F_i'(\varphi(\hat{\omega}^-(m_i)), \varphi(\hat{\omega}^+(m_i)))\} \\
\text{subject to} \\
g_j(y_j, \varphi(\omega^-(o_j)), \varphi(\omega^+(o_j)), z_j) \leq 0, \quad j \in J \\
G_i'(\varphi(\hat{\omega}^-(m_i)), \varphi(\hat{\omega}^+(m_i))) \leq 0, \quad i \in I \\
y_j \in \{0, 1\}, z_j \geq 0, \quad j \in J.
\end{array}\right\} \quad (2)$$

4. PRACTICAL EXAMPLE

The combinatorial part of an industrial instance of PNS is given here. For producing material A61, experimental investigations have given rise to a set of plausible operating units and the set of raw materials. We have set M = {A1, A2, ..., A65} as the set of materials, and R = {A1, A2, A3, A4, A6, A7, A8, A11, A15, A17, A18, A19, A20, A23, A27, A28, A29, A30, A34, A43, A47, A49, A52, A54} as the set of raw materials; set O of plausible operating units is listed in Table I. The maximal structure of this instance is a proper subgraph of (M, O); operating units 14, 18, 30, 34, and 35, and the corresponding materials do not belong to this maximal structure. The maximal structure in Figure 3 was determined by Algorithm MSG [2]. One of the 3465 different solution-structures is illustrated in Figure 4.

5. CONCLUDING REMARKS

A special class of network synthesis problems, the process network synthesis (PNS), has been defined. This class of problems is frequently found in designing industrial systems. The essential properties of the feasible structures of PNS have been summarized, and a MINLP model and a reduced MINLP model for PNS have been developed.

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REFERENCES

[1] F. Friedler, K. Tarjan, Y. W. Huang, and L. T. Fan, Graph theoretic approach to process synthesis: axioms and theorems. *Chem. Eng. Sci.* **47** (1992) 1973–1988.

[2] F. Friedler, K. Tarjan, Y. W. Huang, and L. T. Fan, Combinatorial algorithms for process synthesis. *Comput. Chem. Eng.* **16** (1992) S313–S320.

[3] R. S. H. Mah, *Chemical Process Structures and Information Flows,* Butterworths, Stoneham, MA (1990).

[4] M. Minoux, Network synthesis and optimum network design problems: models, solution methods and applications. *Networks* **19** (1989) 313–360.