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GRAPH-THEORETIC APPROACH TO PROCESS SYNTHESIS: AXIOMS AND THEOREMS

F. FRIEDLER
Department of Systems Engineering, Research Institute for Technical Chemistry, Hungarian Academy of Sciences, Veszprém, P.F. 125, 8201, Hungary

K. TARJÁN
Institute of Mathematics, University of Veszprém, Veszprém, Hungary

and

Y. W. HUANG† and L. T. FAN
Department of Chemical Engineering, Kansas State University, Manhattan, KS 66506, U.S.A.

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Abstract—An innovative approach, based on both graph theory and combinatorial techniques, has been proposed for facilitating the synthesis of a process system. In contrast to other general purpose mathematical programming methods, this innovative approach is designed to cope with the specificities of a process system: it represents the structure of a process system by a unique bipartite graph, termed a P-graph, and captures not only the syntactic but also the semantic contents of the process system. An axiom system underlying the approach has been constructed to define the combinatorially feasible process structures. This axiom system is based on a given set of specifications for the process synthesis problem. Such specifications include the types of operating units and the raw materials, products, by-products, and a variety of waste associated with these operating units. All feasible structures of the process system are embedded in the maximal structure, from which individual solution-structures can be extracted subject to various technical, environmental, economic, and societal constraints. Various theorems have been derived from the axiom system to ensure that this approach is mathematically rigorous, thereby rendering it possible to develop efficient process synthesis methods on the basis of a rigorous mathematical foundation. Examples are presented to highlight the significance and efficacy of the present approach.

INTRODUCTION

A number of methods have been developed for the synthesis of process systems, or process synthesis in brief. These methods can be classified according to whether they are based purely on heuristics or algorithms, i.e. mathematical programming methods. Nevertheless, some methods contain both heuristics and algorithms. Such methods can be classified as heuristic-oriented if they depend more heavily on the former than on the latter, and as algorithmic-oriented if they depend more heavily on the latter than on the former.

A recent review indicates that methods applied to the synthesis of industrial processes are mainly heuristic or heuristic-oriented (Westerberg, 1989). This trend is favored by some practitioners and researchers of process synthesis; however, a concomitant development of mathematical programming methods is highly desirable. It was observed as early as 1975 that the mathematical system theory had not flourished in process synthesis (Westerberg and Stephanopoulos, 1975); it appears that hitherto no mathematical tool has been available to analyze the theoretical aspects of process synthesis, which, from the mathematical point of view, belongs to a special class of network synthesis problems. Nevertheless, since its mathematical foundation has not been established, the recent review article on the network synthesis methods (Minoux, 1989) does not cite even a single publication from the chemical engineering literature.

Efforts to apply the mathematical programming methods, e.g. mixed integer nonlinear programming (MINLP) to various process synthesis problems have produced encouraging results [e.g. Floudas *et al.* (1989) and Kocis and Grossmann (1989)]. Nevertheless, a number of theoretical questions remain to be resolved before an automatic synthesis of a large industrial process can be realized.

It is often arduous to solve a large process synthesis problem by a mathematical programming method, e.g. MINLP, which supposedly ensures the global optimality of the resultant solution. This can be attributed to the fact that the computational difficulty almost always increases exponentially with the size of the problem. A possible approach for obviating this difficulty is to simplify the model of the problem by additional mathematical methods [e.g. Papadimitriou and Steiglitz (1982) and Nemhauser and Wolsey (1988)]. Unfortunately, these methods do not exploit the peculiar characteristics of the MINLP model of the synthesis problem; thus, they are unnecessarily complex, while the size of each solvable problem is rather small in terms of discrete variables.

†Also affiliated with Odin Corporation, Manhattan, KS 66502, U.S.A.

Another possible approach for enhancing the effectiveness of a mathematical programming method is to exploit the unique features of the structure of the process to be synthesized in the earliest possible stage of solution. This approach will be attempted in the present work; it is analogous to the recent trend in discrete optimization to exploit the combinatorial structure of the problem [e.g. Tardos (1990)].

If unique features of a process structure are formulated into a complete mathematical axiom system, the search for the optimal structure can be restricted to the set of feasible structures in process synthesis. Furthermore, this axiom system would allow the development of a variety of algorithms for the manipulating process structures explicitly; the validity of all these algorithms can be proved rigorously. In addition, the axiom system would render it possible to establish the principles for structure decomposition or simplification, to apply combinatorial enumeration, to determine the number of feasible structures for the process, and to generate algorithmically the "super-structure" of the problem.

The approach proposed in the present work is mathematically innovative; it is based on the concept underlying a combinatorial algorithm for process synthesis developed earlier (Frieldler *et al.*, 1979; Friedler and Pintér, 1988). This original concept is formalized in the present work, thereby giving rise to the definition of the process graph (P-graph) to facilitate the process synthesis. The present approach relies heavily on both graph theory and combinatorial techniques. It focuses on structures of the process system and rigorously examines such structures from the mathematical perspective. An axiom system underlying the proposed approach is constructed, and various theorems are derived from it to ensure that the rigorous mathematical basis for process synthesis be established through the present approach. Though the present approach is general, the focus is on its application to the first steps of process design, namely, process development, planning and basic design (Umeda, 1983).

The problem of structure representation in process synthesis is examined at the outset. This is followed by the definitions of the set of feasible process structures by an axiom system. The theorems on the properties of these feasible structures are presented; this leads to a formal definition and properties of a maximal structure in which all feasible structures are embedded. Individual solution-structures can be extracted from the maximal structure subject to various technical, economic, environmental, and societal constraints.

STRUCTURAL REPRESENTATION

The foundation of the present approach comprises the graph of a new type for effective structural representation of a process system. This graph has been proposed to alleviate difficulties encountered by approaches based on conventional graphs, e.g. digraph and signal-flow graph. In the digraph representation of a process system, the operating units correspond to the vertices, and the connections to the arcs of the graph. In the signal-flow graph representation of a process system, the vertices of the graph are associated with the materials of the process. While these conventional graphs are suitable for representing and analyzing a process system [e.g. Mah (1983, 1990) and Dudczak (1986)], they are not suitable for process synthesis as demonstrated in the following simple examples.

Example 1

Cases (1.1) and (1.2) described below can be represented by the same digraph shown in Fig. 1.

Case (1.1). Two different materials are produced separately, one by operating unit 02 and the other by operating unit 03. Moreover, it is necessary to feed both of these materials to operating unit 01 to generate the final product.

Case (1.2). One material is produced by both operating units 02 and 03. This material is subsequently fed to operating unit 01 to generate the final product.

Note that while both operating units 02 and 03 are necessary to produce the product in the first case, either unit is sufficient in the second case.

Example 2

Cases (2.1) and (2.2) described below are represented by the same signal-flow graph of Fig. 2.

Case (2.1). Two separate operating units, one receiving material B as its input and the other material C as its input, produce the same material which is subsequently fed to another operating unit where material A (product) is generated.

Case (2.2). A single operating unit, receiving materials B and C as its inputs, produces a material which is subsequently fed to another operating unit where material A (product) is generated.

Note that while the first case requires three operating units, the second case requires only two units. Obviously, the semantics, i.e. meaning, of the figure is

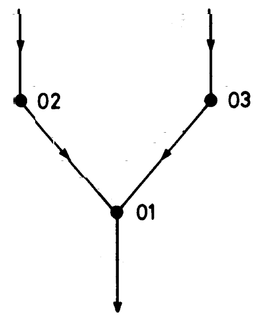

Fig. 1. Digraph: note that it is incapable of uniquely characterizing a synthesis problem as demonstrated by example 1.

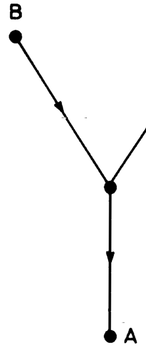

Fig. 2. Signal-flow graph: note that it is incapable of uniquely characterizing a synthesis problem as demonstrated by example 2.

unclear. As in Fig. 1, Fig. 2 fails to describe the process system in clear semantics.

Examples 1 and 2 demonstrate that neither of the two most popular conventional graphs is semantically rich enough to faithfully represent a process structure. The semantics of a process structure is concerned with the meaning of individual materials and operating units and the relationship between them, while the syntax of the process structure is concerned with the ordered organization of the flow of the materials and the operating units.

Both a digraph and a signal-flow graph can orderly encode a process structure into a graph representation. However, as demonstrated by the examples above, the former is not sufficient to uniquely represent individual materials and their relationship, and the latter is not sufficient to uniquely represent individual operating units and their relationship. Hence, a graph more sophisticated than a conventional one, such as the digraph or signal-flow graph, is required to uniquely characterize a synthesis problem. For this reason, a special graph is introduced to capture not only the syntactic but also the semantic contents of the process structure.

Basic terminology

Let $\mathscr{M}$ be a given nonempty finite set of all materials which are to be involved in the synthesis of a process system; it may be a set of names or a set of vectors of characteristics of these materials. The exact description, i.e. the contents, of set $\mathscr{M}$ may vary depending on the level of precision or detail desired. While a coarse description of the materials may suffice for approximate presentation of the system, a fine description may be required for detailed presentation. For example, both

$$\mathscr{M}_1 = \{x_1, x_2, x_3, x_4, x_5, x_6\}$$

and

$$\mathscr{M}_2 = \{(A, \varnothing, \varnothing), (\varnothing, B, \varnothing), (\varnothing, \varnothing, C), (A, B, \varnothing), (\varnothing, B, C), (A, B, C)\}$$

may represent the materials involved in a sharp separation scheme of a mixture of components A, B, and C. Obviously, the latter is more descriptive than the former although both may contain identical information.

A synthesis problem involving materials represented by set $\mathscr{M}$ can be defined by triplet ($\mathscr{P}$, $\mathscr{R}$, $\mathscr{O}$) where $\mathscr{P}$ is the set of products, $\mathscr{R}$, the set of raw materials, and $\mathscr{O}$, the set of operating units for the problem. The relationships among $\mathscr{M}$, $\mathscr{P}$, $\mathscr{R}$, and $\mathscr{O}$ can be mathematically expressed as follows:

$$\mathscr{P} \subset \mathscr{M}, \mathscr{R} \subset \mathscr{M}, \quad \text{and} \quad \mathscr{R} \cap \mathscr{P} = \varnothing \tag{1}$$

and

$$\mathscr{O} \subseteq \wp(\mathscr{M}) \times \wp(\mathscr{M}) \tag{2}$$

where $\wp(*)$ is a power set. Usually, if $(\alpha, \beta) \in \mathscr{O}$, then α designates the set of input materials, and β the set of output materials of operating unit (α, β). If waste is to be treated in the process being synthesized, α designates the set of materials with potential loss, and β the set of materials with potential gain with respect to the operating unit. A material is considered to be a "potential loss" when it may eventually contribute negatively to the objective function of the overall process being synthesized, e.g. profit; otherwise, it is considered to be a "potential gain". A typical example of the former is a raw material, and that of the latter is a final product. Note that waste, by definition, contributes negatively to the synthesized process. It is, therefore, a potential loss.

The definition of $\mathscr{M}$, $\mathscr{P}$, $\mathscr{R}$, and $\mathscr{O}$ will be illustrated with the sharp separation of a mixture of components A, B, and C, i.e. mixture ABC, by distillation described in the preceding paragraph. Suppose that the volatilities of these components are ranked in the descending order of A, B, and C. Then, for this example, we have

$$\mathscr{M}_1 = \{x_1, x_2, x_3, x_4, x_5, x_6\}$$
$$\mathscr{P}_1 = \{x_1, x_2, x_3\}$$
$$\mathscr{R}_1 = \{x_6\}$$

and

$$\mathscr{O}_1 = \{(\{x_4\}, \{x_1, x_2\}), (\{x_5\}, \{x_2, x_3\}), (\{x_6\}, \{x_1, x_5\}), (\{x_6\}, \{x_3, x_4\})\}.$$

Naturally,

$$\mathscr{O}_1 \subseteq \wp(\mathscr{M}_1) \times \wp(\mathscr{M}_1)$$

with

$$\wp(\mathscr{M}_1) = \{\varnothing, \{x_1\}, \{x_2\}, \{x_3\}, \{x_4\}, \{x_5\}, \{x_6\}, \{x_1, x_2\}, \{x_1, x_3\}, \ldots, \{x_1, x_2, x_3, x_4, x_5, x_6\}\}$$

and

$$\wp(\mathscr{M}_1) \times \wp(\mathscr{M}_1) = \{(\varnothing, \varnothing), (\varnothing, \{x_1\}), \ldots, (\varnothing, \{x_1, x_5\}), (\varnothing, \{x_1, x_6\}), \ldots, (\varnothing, \{x_1, x_2, x_3, x_4, x_5, x_6\}),$$

$(\{x_1\}, \varnothing), (\{x_1\}, \{x_1\}), \ldots, (\{x_1\}, \{x_1, x_5\}),$
$(\{x_1\}, \{x_1, x_6\}), \ldots, (\{x_1\},$
$\{x_1, x_2, x_3, x_4, x_5, x_6\}),$

$(\{x_6\}, \varnothing), (\{x_6\}, \{x_1\}), \ldots, (\{x_6\}, \{x_1, x_5\}),$
$(\{x_6\}, \{x_1, x_6\}), \ldots,$
$(\{x_1\}, \{x_1, x_2, x_3, x_4, x_5, x_6\}),$

.

$(\{x_1, x_2, x_3, x_4, x_5, x_6\}, \varnothing),$
$(\{x_1, x_2, x_3, x_4, x_5, x_6\},$
$\{x_1\}), \ldots, (\{x_1, x_2, x_3, x_4, x_5, x_6\},$
$\{x_1, x_5\}), (\{x_1, x_2, x_3, x_4, x_5, x_6\},$
$\{x_1, x_6\}), \ldots, (\{x_1, x_2, x_3, x_4, x_5, x_6\},$
$\{x_1, x_2, x_3, x_4, x_5, x_6\})\}.$

Accordingly,

$$(\{x_6\}, \{x_1, x_5\})$$

or

$$(\{(A, B, C)\}, \{(A, \varnothing, \varnothing), (\varnothing, B, C)\})$$

based on $\mathscr{M}_1$ or $\mathscr{M}_2$, represents an operating unit separating mixture *ABC* into component *A* and mixture *BC* in the separation scheme, respectively (see Fig. 3).

Process graph (P-graph)

As demonstrated, conventional graphs are incapable of uniquely representing process structures in synthesis. Thus, a graph containing additional details is required. To represent a process structure in synthesis, it is possible to adopt a directed bipartite graph (Friedler *et al.*, 1979; Friedler and Pintér, 1988) or to complete a digraph representation by fictitious (the so-called interconnection) vertices (Grossmann, 1989). The bipartite graph representation is preferred here for the unified mathematical description of the process structure. A graph is bipartite if its vertices can be partitioned into two disjoint sets, and no two vertices in the same set are adjacent. This has given rise to the

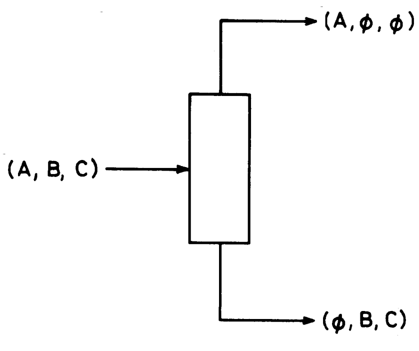

Fig. 3. Operating unit separating mixture *ABC* into component *A* and mixture *BC*.

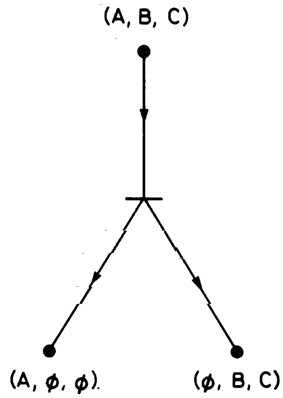

Fig. 4. P-graph representation of Fig. 3.

definition of a process graph, or P-graph in short. Figure 4 is the P-graph representation of the structure of the operating unit separating mixture *ABC* into component *A* and mixture *BC* shown in Fig. 3. In Fig. 4, a material in the process is symbolized by a circle, designating an M-type vertex; an operating unit is symbolized by a horizontal bar, designating an O-type vertex. The mathematical definition of a P-graph and that of a process structure, such as a P-graph, will be elaborated in what follows.

Given two finite sets $\mathscr{m}$ and $\mathscr{o}$ with

$$\mathscr{o} \subseteq \wp(\mathscr{m}) \times \wp(\mathscr{m}) \tag{3}$$

a P-graph is defined to be a pair, $(\mathscr{m}, \mathscr{o})$, as follows:

(i) The vertices of the graph are the elements of

$$\mathscr{V} = \mathscr{m} \cup \mathscr{o}. \tag{4}$$

Those belonging to set $\mathscr{m}$ are M-type vertices, and those belonging to set $\mathscr{o}$ are O-type vertices.

(ii) The arcs of the graph are the elements of

$$\mathscr{A} = \mathscr{A}_1 \cup \mathscr{A}_2 \tag{5}$$

where

$$\mathscr{A}_1 = \{(x, y) \mid y = (\alpha, \beta) \in \mathscr{o} \text{ and } x \in \alpha\} \tag{6}$$

and

$$\mathscr{A}_2 = \{(y, x) \mid y = (\alpha, \beta) \in \mathscr{o} \text{ and } x \in \beta\}. \tag{7}$$

In these expressions, x designates an M-type vertex, y an O-type vertex, α a set of M-type vertices from which arcs are directed into the O-type vertex, and β a set of M-type vertices to which arcs are directed out of the O-type vertex. In other words, each arc in $\mathscr{A}_1$ is from an M-type vertex to an O-type vertex, and that in $\mathscr{A}_2$ from an O-type vertex to an M-type vertex. Note that the arcs are only implicitly defined for the P-graph. This is in sharp contrast to the conventional way of defining a graph where both the vertices and arcs are explicitly given. The advantage of this non-conventional definition will be elaborated later.

In summary, two types of vertices, M- and O-types, exist for P-graph $(\mathscr{m}, \mathscr{o})$; an arc which is directed spans two vertices of different types only. The union and intersection of the two P-graphs, $(\mathscr{m}_1, \mathscr{o}_1)$ and

$(\mathscr{m}_2, \mathscr{o}_2)$, are defined, respectively, as

$$(\mathscr{m}_1, \mathscr{o}_1) \cup (\mathscr{m}_2, \mathscr{o}_2) = (\mathscr{m}_1 \cup \mathscr{m}_2, \mathscr{o}_1 \cup \mathscr{o}_2) \quad (8)$$

and

$$(\mathscr{m}_1, \mathscr{o}_1) \cap (\mathscr{m}_2, \mathscr{o}_2) = (\mathscr{m}_1 \cap \mathscr{m}_2, \mathscr{o}_1 \cap \mathscr{o}_2) \quad (9)$$

both of which remain as P-graphs, since

$$\begin{aligned} \mathscr{o}_1 \cup \mathscr{o}_2 &\subseteq (\wp(\mathscr{m}_1) \times \wp(\mathscr{m}_1)) \cup (\wp(\mathscr{m}_2) \times \wp(\mathscr{m}_2)) \\ &\subseteq \wp(\mathscr{m}_1 \cup \mathscr{m}_2) \times \wp(\mathscr{m}_1 \cup \mathscr{m}_2) \quad (10) \end{aligned}$$

and

$$\begin{aligned} \mathscr{o}_1 \cap \mathscr{o}_2 &\subseteq (\wp(\mathscr{m}_1) \times \wp(\mathscr{m}_1)) \cap (\wp(\mathscr{m}_2) \times \wp(\mathscr{m}_2)) \\ &= \wp(\mathscr{m}_1 \cap \mathscr{m}_2) \times \wp(\mathscr{m}_1 \cap \mathscr{m}_2). \quad (11) \end{aligned}$$

Furthermore, $(\mathscr{m}_1, \mathscr{o}_1)$ is a subgraph of $(\mathscr{m}_2, \mathscr{o}_2)$, i.e.

$$(\mathscr{m}_1, \mathscr{o}_1) \subseteq (\mathscr{m}_2, \mathscr{o}_2)$$

if

$$\mathscr{m}_1 \subseteq \mathscr{m}_2 \quad \text{and} \quad \mathscr{o}_1 \subseteq \mathscr{o}_2.$$

The structure of a process can be defined as a P-graph. For synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$, let $\mathscr{m}$ be a subset of material set $\mathscr{M}$, and $\mathscr{o}$ be a subset of operating unit set $\mathscr{O}$. Furthermore, let us suppose that sets $\mathscr{m}$ and $\mathscr{o}$ satisfy formula (3). Then, the structure of the system with set $\mathscr{m}$ of materials and set $\mathscr{o}$ of operating units is formally defined as P-graph $(\mathscr{m}, \mathscr{o})$. This definition leads to the following properties.

(i) Materials and operating units are represented by vertices of the M- and O-types, respectively.
(ii) Two vertices, one of the M-type and the other of the O-type, are linked by an arc if and only if the corresponding linkage is realized in the process represented by the graph.
(iii) An arc originates from the node representing a material with a potential loss towards that representing an operating unit, or from the node representing an operating unit towards that representing a material with a potential gain. This property indicates that the directions of arcs are usually identical to the directions of material flows in a process.

A P-graph can capture not only the syntactic but also the semantic contents of a process system. For the two examples presented at the outset of this paper, three different P-graphs can be constructed to uniquely represent the four cases. Note that cases (1.2) and (2.1), which are identical, are uniquely represented by the P-graph in Fig. 5, case (2.2) in Fig. 6, and case (1.1) in Fig. 7. Another example, a structure of sharp separation of mixture *ABC* into its three components, *A*, *B*, and *C*, can be represented by the P-graphs given in Fig. 8.

Note that in practice, no arbitrary P-graph can represent the structure of a process. Any P-graph must satisfy a set of combinatorial properties of the process, comprising the materials and operating units, to appropriately represent a feasible process structure.

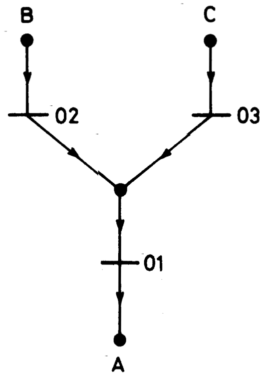

Fig. 5. P-graph uniquely representing cases (1.2) and (2.1).

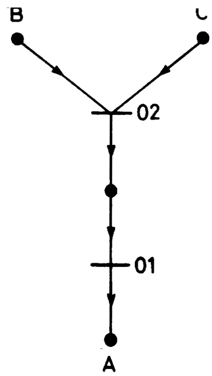

Fig. 6. P-graph uniquely representing case (2.2).

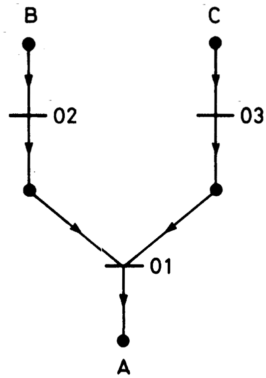

Fig. 7. P-graph uniquely representing case (1.1).

These properties give rise to a set of axioms which are discussed in what follows.

SOLUTION-STRUCTURES

The materials and operating units in a feasible process structure must always conform to certain combinatorial properties. For example, a structure containing no linkage between a raw material and a final product is unlikely to represent any practical process. Hence, it is of vital importance to identify the

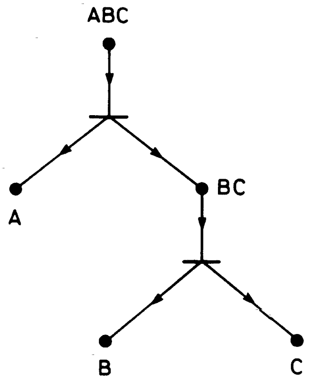

Fig. 8. P-graph representation of a process structure involving sharp separation of mixture ABC into its three components.

general combinatorial properties to which a process structure must conform. More importantly, the properties identified should be satisfied by the structure of any feasible solution of the synthesis problem. Conversely, any P-graph that satisfies these properties can usually be the structure of a feasible solution. In other words, those and only those structures satisfying these properties can be feasible structures of a process: no other structures or constraints need be considered in synthesizing the process.

Axioms

The following set of axioms has been constructed to express the necessary and sufficient combinatorial properties to which a feasible process structure should conform:

- (S1) Every final product is represented in the graph.
- (S2) A vertex of the M-type has no input if and only if it represents a raw material.
- (S3) Every vertex of the O-type represents an operating unit defined in the synthesis problem.
- (S4) Every vertex of the O-type has at least one path leading to a vertex of the M-type representing a final product.
- (S5) If a vertex of the M-type belongs to the graph, it must be an input to or output from at least one vertex of the O-type in the graph.

If a P-graph of a given synthesis problem, $(\mathscr{P}, \mathscr{R}, \mathscr{O})$, satisfies these axioms, it is defined to be a solution-structure of the problem. In other words, for synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$, axiom (S1) demands that all the final products, $\mathscr{P}$, be generated by the process represented by a solution-structure; axiom (S2) explicates the meaning of raw materials, $\mathscr{R}$, which, by definition, should not be generated by the process under consideration; according to axiom (S3), only those operating units, $\mathscr{O}$, that are defined in the problem can appear in a solution-structure; axiom (S4) disallows the existence of an operating unit which is not contributing to product generation; and, according to axiom (S5), only those materials that belong to at least one operating unit of the structure can exist in the structure. The formal mathematical definitions of these axioms are given in Appendix A.

If a vertex of the O-type belongs to a P-graph representing a solution-structure, all materials input to and output from it also belong to the graph according to formula (3), since a solution-structure is required to be a P-graph. Figure 9 depicts an example of two solution-structures for synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$ with

$$\mathscr{M}_3 = \{A, B, C, D, E, F, G, H, I\}$$

$$\mathscr{P}_3 = \{A\}$$

$$\mathscr{R}_3 = \{D, F, H\}$$

and

$$\mathscr{O}_3 = \{(\{C\}, \{A, I\}), (\{B\}, \{A, E\}), (\{D, E\}, \{B\}), (\{E, F\}, \{B\}), (\{F, G\}, \{C\}), (\{H, I\}, \{G\})\}.$$

Note that a solution-structure does not necessarily contain all the components defined in the set of materials, e.g. $\mathscr{M}_3$; neither does it necessarily utilize all the components specified in the set of raw materials, e.g. $\mathscr{R}_3$.

Since the final product, A, is present as an M-type vertex in both Fig. 9(a) and (b), axiom (S1) is satisfied by the solution-structures depicted in these figures. Axiom (S2) is satisfied in that vertex F in Fig. 9(a) and vertices F and H in Fig. 9(b) are the only vertices without an input; they represent raw materials. Figure 9(a) contains two operating units, $(\{E, F\}, \{B\})$ and $(\{B\}, \{A, E\})$, and Fig. 9(b) contains three operating units, $(\{H, I\}, \{G\})$, $(\{F, G\}, \{C\})$, and $(\{C\}, \{A, I\})$; all these operating units are defined in the synthesis problem, thereby satisfying axiom (S3). In conformity with axiom (S4), every vertex of the O-type in either

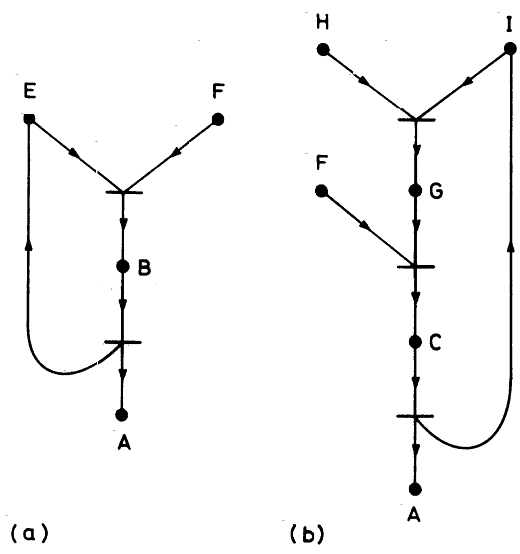

Fig. 9. Two solution-structures for synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$.

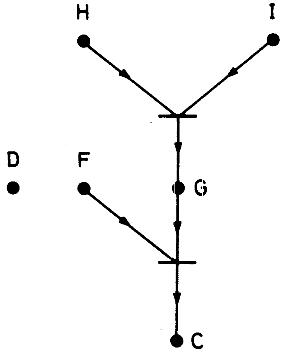

Fig. 10. P-graph that is not a solution-structure for synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$.

Fig. 9(a) or (b) does have at least one path leading to vertex A representing the final product. For example, the path in Fig. 9(a), comprising three arcs, namely, $((\{E, F\}, \{B\}), B)$, $(B, (\{B\}, \{A, E\}))$, and $((\{B\}, \{A, E\}), A)$, links vertex $(\{E, F\}, \{B\})$, representing an operating unit, to vertex A which is the final product. Axiom (S5) is satisfied by virtue of the fact that every vertex of the M-type belonging to the graph of either Fig. 9(a) or (b) is an input to or output from at least one vertex of the O-type in the respective graph. Thus, all axioms are satisfied by the structures in Fig. 9(a) and (b). As a counterexample, Fig. 10 illustrates a P-graph that is not a solution-structure of synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$, because axioms (S1), (S2), (S4), and (S5) are not satisfied.

Theorems

Let $S(\mathscr{P}, \mathscr{R}, \mathscr{O})$ be the set of all solution-structures for process synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$. The following theorems are derived on S. The proofs of these theorems are given in Appendix B.

Theorem 1 (closure under union on solution-structures): *The union of two solution-structures remains a solution-structure, that is, if*

$$\sigma_1 \in S(\mathscr{P}, \mathscr{R}, \mathscr{O}) \quad \text{and} \quad \sigma_2 \in S(\mathscr{P}, \mathscr{R}, \mathscr{O})$$

then

$$(\sigma_1 \cup \sigma_2) \in S(\mathscr{P}, \mathscr{R}, \mathscr{O}).$$

In general, however, $(\sigma_1 \cap \sigma_2)$ *will not be an element of* $S(\mathscr{P}, \mathscr{R}, \mathscr{O})$.

Theorem 2 (composition/decomposition of solution-structures): *Solution-structures generating different products can be composed to yield a solution-structure generating all the products, that is, if*

$$\sigma_1 \in S(\mathscr{P}_1, \mathscr{R}, \mathscr{O}) \quad \text{and} \quad \sigma_2 \in S(\mathscr{P}_2, \mathscr{R}, \mathscr{O})$$

then

$$(\sigma_1 \cup \sigma_2) \in S(\mathscr{P}_1 \cup \mathscr{P}_2, \mathscr{R}, \mathscr{O}).$$

Conversely, a solution-structure generating a large set of products can be decomposed to yield solution-structures generating subsets of products. In other words, if

$$\sigma \in S(\mathscr{P}_1 \cup \mathscr{P}_2, \mathscr{R}, \mathscr{O})$$

then there exist σ_1 *and* σ_2 *such that*

$$\sigma_1 \in S(\mathscr{P}_1, \mathscr{R}, \mathscr{O}), \sigma_2 \in S(\mathscr{P}_2, \mathscr{R}, \mathscr{O}), \text{ and } \sigma = (\sigma_1 \cup \sigma_2).$$

Theorem 3 (extraction of a solution-structure based on common products): *A solution-structure generating a specific set of products can be extracted from the union of the solution-structures generating this set of products as common products. In other words, if*

$$\sigma_1 \in S(\mathscr{P}_1, \mathscr{R}, \mathscr{O}), \sigma_2 \in S(\mathscr{P}_2, \mathscr{R}, \mathscr{O}), \quad \text{and} \quad \mathscr{P}_1 \cap \mathscr{P}_2 \neq \emptyset$$

then there exists

$$\sigma_3 \in S(\mathscr{P}_1 \cap \mathscr{P}_2, \mathscr{R}, \mathscr{O})$$

such that

$$\sigma_3 \subseteq (\sigma_1 \cup \sigma_2).$$

Theorem 4 (extensibility of solution-structures): *A solution-structure generating a smaller set of products can be extended so that it is contained in a solution-structure generating a larger set of products provided that the smaller set is a subset of the larger set. In other words, if*

$$\mathscr{P}_1 \subset \mathscr{P}_2, \sigma_1 \in S(\mathscr{P}_1, \mathscr{R}, \mathscr{O}), \text{ and } S(\mathscr{P}_2, \mathscr{R}, \mathscr{O}) \neq \emptyset$$

then there exists

$$\sigma_2 \in S(\mathscr{P}_2, \mathscr{R}, \mathscr{O})$$

such that

$$\sigma_1 \subseteq \sigma_2$$

Theorem 5 (contractibility of solution-structures): *A solution-structure generating a larger set of products can be contracted to yield a solution-structure generating a smaller nonempty set of products provided that the smaller set is a subset of the larger set. In other words, if*

$$\emptyset \neq \mathscr{P}_1 \subset \mathscr{P}_2 \quad \text{and} \quad \sigma_2 \in S(\mathscr{P}_2, \mathscr{R}, \mathscr{O})$$

then there exists

$$\sigma_1 \in S(\mathscr{P}_1, \mathscr{R}, \mathscr{O})$$

such that

$$\sigma_1 \subseteq \sigma_2.$$

Theorem 6 (operating-unit-set expansibility of solution-structures): *A solution-structure of a synthesis problem involving a smaller set of operating units is also a solution-structure of a synthesis problem involving a larger set of operating units provided that the smaller set is a subset of the larger set. In other words, if*

$$\mathscr{O}_1 \subset \mathscr{O}_2 \quad \text{and} \quad \sigma \in S(\mathscr{P}, \mathscr{R}, \mathscr{O}_1)$$

then

$$\sigma \in S(\mathscr{P}, \mathscr{R}, \mathscr{O}_2)$$

i.e.

$$S(\mathscr{P}, \mathscr{R}, \mathscr{O}_1) \subseteq S(\mathscr{P}, \mathscr{R}, \mathscr{O}_2).$$

Examples

The theorems derived are elaborated further through synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$ defined previously, and synthesis problem $(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$ specified by

$$\mathscr{M}_4 = \{A, B, C, D, E, F, G, H, I, J, K, L, M, N, O, P, R, S\}$$

$$\mathscr{P}_4 = \{A, B, C\}$$

$$\mathscr{R}_4 = \{G, N, O, P, R\}$$

and

$$\mathscr{O}_4 = \{(\{D\},\{A\}),(\{E\},\{B\}),(\{I\},\{C\}),(\{F\},\{D,K\}),(\{F,G\},\{E,K\}),(\{H\},\{E,S\}),(\{K\},\{F\}),(\{K,L\},\{F\}),(\{L,M\},\{H,I\}),(\{M,N\},\{I\}),(\{O\},\{K\}),(\{P,R,S\},\{L,M\}),(\{J,K\},\{F\})\}.$$

Theorem 1 asserts that the set of solution-structures is closed under union. For example, the P-graph in Fig. 11 is the union of the two P-graphs, i.e. two solution-structures, of synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$ in Fig. 9. Clearly, it is also a solution-structure for the problem. In contrast, it should be obvious that the intersection of two solution-structures usually does not yield a solution-structure.

Theorem 2 states that for synthesis problems with common sets of raw materials and operating units, a solution-structure representing a process generating product set $\mathscr{P}(=\mathscr{P}_1 \cup \mathscr{P}_2)$ can be composed through the union of two solution-structures, one representing the process generating product set $\mathscr{P}_1$ and the other product set $\mathscr{P}_2$. Conversely, the solution-structure representing the process generating product set $\mathscr{P}$ can be decomposed into two solution-structures, one representing the process generating product set $\mathscr{P}_1$ and the other product set $\mathscr{P}_2$. For synthesis problem $(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$, Fig. 12 contains a solution-structure which is the union of two solution-structures. The graph, consisting of region I and the shaded region, is a solution-structure for synthesis problem $(\mathscr{P}'_4, \mathscr{R}_4, \mathscr{O}_4)$, and the graph, consisting of region II and the shaded region, is a solution-structure for synthesis problem $(\mathscr{P}''_4, \mathscr{R}_4, \mathscr{O}_4)$. Note that

$$\mathscr{P}'_4 = \{A, B\}$$

$$\mathscr{P}''_4 = \{B, C\}$$

and

$$\mathscr{P}'_4 \cup \mathscr{P}''_4 = \{A, B, C\} = \mathscr{P}_4.$$

Similarly, theorem 3 states that for two synthesis problems with common sets of raw materials and operating units, a third solution-structure can be extracted from the union of two solution-structures, σ_1 and σ_2, provided that they represent, respectively, two processes generating product sets $\mathscr{P}_1$ and $\mathscr{P}_2$ which intersect. This third solution-structure represents a process generating product set $\mathscr{P}(=\mathscr{P}_1 \cap \mathscr{P}_2)$. In Fig. 12, the graph in the shaded area is a solution-structure for synthesis problem $(\mathscr{P}'''_4, \mathscr{R}_4, \mathscr{O}_4)$ where

$$\mathscr{P}'''_4 = \{B\}.$$

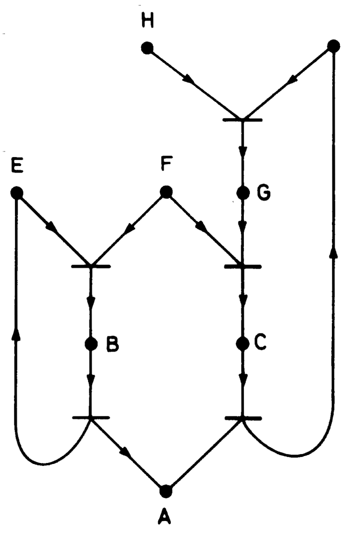

Fig. 11. Solution-structure for synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$, which has been generated through the union of the two solution-structures of Fig. 9.

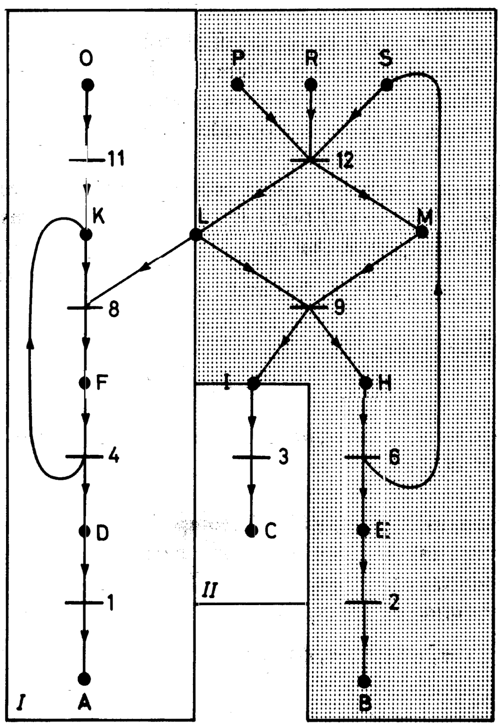

Fig. 12. Composition and decomposition of the solution-structures of synthesis problem $(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$.

This is a subgraph of the union of the graphs representing solution-structures for problems $(\mathscr{P}'_4, \mathscr{R}_4, \mathscr{O}_4)$ and $(\mathscr{P}''_4, \mathscr{R}_4, \mathscr{O}_4)$. Clearly,

$$\mathscr{P}'_4 \cap \mathscr{P}''_4 = \{B\} = \mathscr{P}'''_4.$$

Theorem 4 explicates that a solution-structure of a smaller synthesis problem, $(\mathscr{P}_1, \mathscr{R}, \mathscr{O})$, can readily be extended so that it is contained in a solution-structure of a larger synthesis problem, $S(\mathscr{P}_2, \mathscr{R}, \mathscr{O})$, where $\mathscr{P}_1 \subset \mathscr{P}_2$. Conversely, a solution-structure of the larger problem can be contracted to yield a solution-structure for the smaller problem according to theorem 5. As an example for extensibility of solution-structures, the graph in the shaded region of Fig. 12 is a solution-structure for synthesis problem $(\mathscr{P}'''_4, \mathscr{R}_4, \mathscr{O}_4)$. It can be extended to incorporate region I, thereby generating a solution-structure for synthesis problem $(\mathscr{P}'_4, \mathscr{R}_4, \mathscr{O}_4)$, where

$$\mathscr{P}'''_4(=\{B\}) \subset \mathscr{P}'_4(=\{A, B\}).$$

Theorem 6 claims that for synthesis problems with common sets of products and raw materials, a solution-structure of the synthesis problem with the smaller set of operating units is also a solution-structure of that with the larger set of operating units. For example, Fig. 12 in its entirety is a solution-structure, which is an element of

$$S(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4 \backslash \{(\{K\}, \{F\})\})$$

since operating unit $(\{K\}, \{F\})$ does not appear in the structure. Naturally, the solution-structure of Fig. 12 is also an element of $S(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$.

MAXIMAL STRUCTURES

Since the set of solution-structures for process synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$ is finite and closed under the union operation according to theorem 1, it contains an element, $\mu(\mathscr{P}, \mathscr{R}, \mathscr{O})$, which is the union of all its elements, i.e.

$$\mu(\mathscr{P}, \mathscr{R}, \mathscr{O}) = \bigcup_{\sigma \in S(\mathscr{P}, \mathscr{R}, \mathscr{O})} \sigma \qquad (12)$$

provided that $S(\mathscr{P}, \mathscr{R}, \mathscr{O}) \neq \varnothing$. Then, $\mu(\mathscr{P}, \mathscr{R}, \mathscr{O})$ is termed the maximal structure of synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$. In this graph, each arc and vertex belongs to at least one solution-structure, and each solution-structure is a subgraph. A structure corresponding to the maximal structure is often referred to as a "super-structure" in the literature of process synthesis [e.g. Floudas (1987)]. It appears that eq. (12) represents the first formal definition of a super-structure which hitherto has been unavailable. The term "maximal structure" is adopted for two reasons. First, it is more expressive semantically than the super-structure. Second, the term super-structure has a different connotation in computer science [e.g. Cantone *et al.* (1989)]. The maximal structure for the previously defined synthesis problem, $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$, is depicted in Fig. 13, and that for synthesis problem $(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$ in Fig. 14. The maximal structure of separation problem

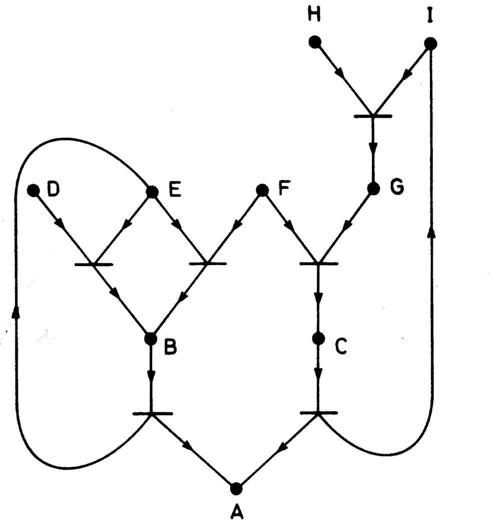

Fig. 13. Maximal structure of synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$.

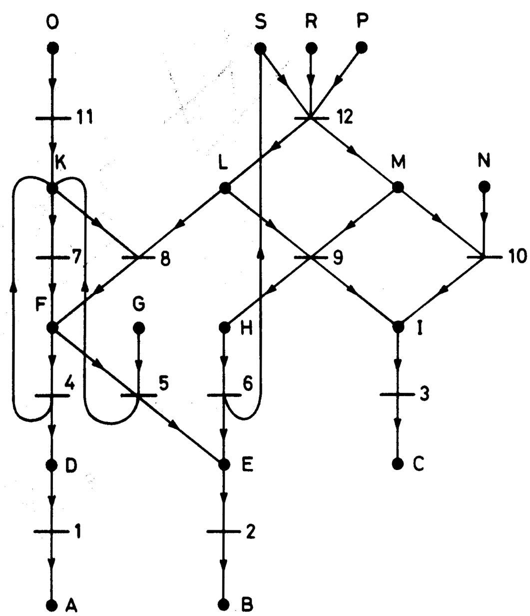

Fig. 14. Maximal structure of synthesis problem $(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$.

C6 (Rodrigo and Seader, 1975; Nath and Motard, 1981) is given in Fig. 15, and the maximal structure of the synthesis problem of Grossmann (1985) is given in Fig. 16.

If pair $(\mathscr{M}, \underline{\mathscr{O}})$ represents the P-graph of $\mu($

then $\underline{\mathscr{O}}$ must represent a set of operating u

which

$$\underline{\mathscr{O}} \subseteq \mathscr{O}$$

and

$$S(\mathscr{P}, \mathscr{R}, \mathscr{O}) = S(\mathscr{P}, \mathscr{R}, \underline{\mathscr{O}})$$

and $\mathscr{M}$ represents the set of materials associa

$\underline{\mathscr{O}}$. Stated differently, not all operating units

$\mathscr{R}, \mathscr{O}$),

nits, for

(13)

(14)

ted with

given in

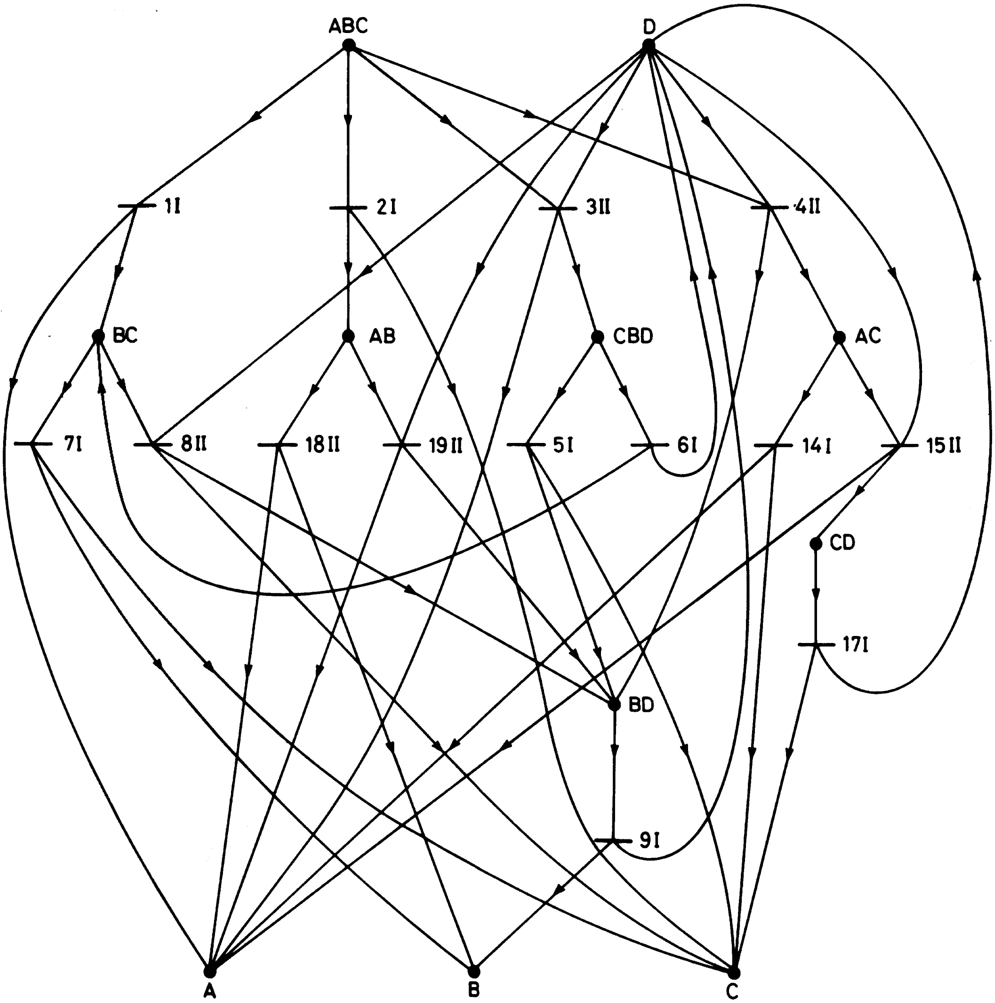

Fig. 15. Maximal structure of separation problem C6 (Rodrigo and Seader, 1975; Nath and Motard, 1981).

the specifications of synthesis problem $(\mathcal{P}, \mathcal{R}, \mathcal{O})$ need appear in the maximal structure of the problem. The maximal structure contains a minimal subset of $\mathcal{O}$, $\underline{\mathcal{O}}$, for which

$$\mu(\mathcal{P}, \mathcal{R}, \mathcal{O}) = \mu(\mathcal{P}, \mathcal{R}, \underline{\mathcal{O}}). \tag{15}$$

For example, in synthesis problem $(\mathcal{P}_4, \mathcal{R}_4, \mathcal{O}_4)$, for a subset of $\mathcal{O}_4$, $\underline{\mathcal{O}}_4$, where

$$\underline{\mathcal{O}}_4 = \mathcal{O}_4 \backslash \{(\{J, K\}, \{F\})\} \subseteq \mathcal{O}$$

we find that

$$\mu(\mathcal{P}_4, \mathcal{R}_4, \mathcal{O}_4) = \mu(\mathcal{P}_4, \mathcal{R}_4, \underline{\mathcal{O}}_4)$$

and

$$S(\mathcal{P}_4, \mathcal{R}_4, \mathcal{O}_4) = S(\mathcal{P}_4, \mathcal{R}_4, \underline{\mathcal{O}}_4).$$

It should be noted that an arbitrary subset of $\mathcal{O}$ does not necessarily satisfy eq. (15).

Theorems

Let $\mu(\mathcal{P}, \mathcal{R}, \mathcal{O})$ be the maximal structure for process synthesis problem $(\mathcal{P}, \mathcal{R}, \mathcal{O})$. The following theorems are derived on μ; see Appendix B for proofs.

Theorem 7 (maximal structure as a solution-structure): *The maximal structure is one of the solution-structures, that is,*

$$\mu(\mathcal{P}, \mathcal{R}, \mathcal{O}) \in S(\mathcal{P}, \mathcal{R}, \mathcal{O}).$$

Theorem 8 (composition of maximal structures): *Maximal structures generating different products can be composed to yield a maximal structure generating all the products, that is,*

$$\mu(\mathcal{P}_1 \cup \mathcal{P}_2, \mathcal{R}, \mathcal{O}) = \mu(\mathcal{P}_1, \mathcal{R}, \mathcal{O}) \cup \mu(\mathcal{P}_2, \mathcal{R}, \mathcal{O}).$$

Theorem 9 (extraction of a maximal structure): *A maximal structure generating a smaller set of products can be extracted from a maximal structure generating a larger set of products provided that the smaller set is a subset of the larger set. In other words, if*

$$\mathcal{P}_1 \subseteq \mathcal{P}_2$$

then

$$\mu(\mathcal{P}_1, \mathcal{R}, \mathcal{O}) \subseteq \mu(\mathcal{P}_2, \mathcal{R}, \mathcal{O}).$$

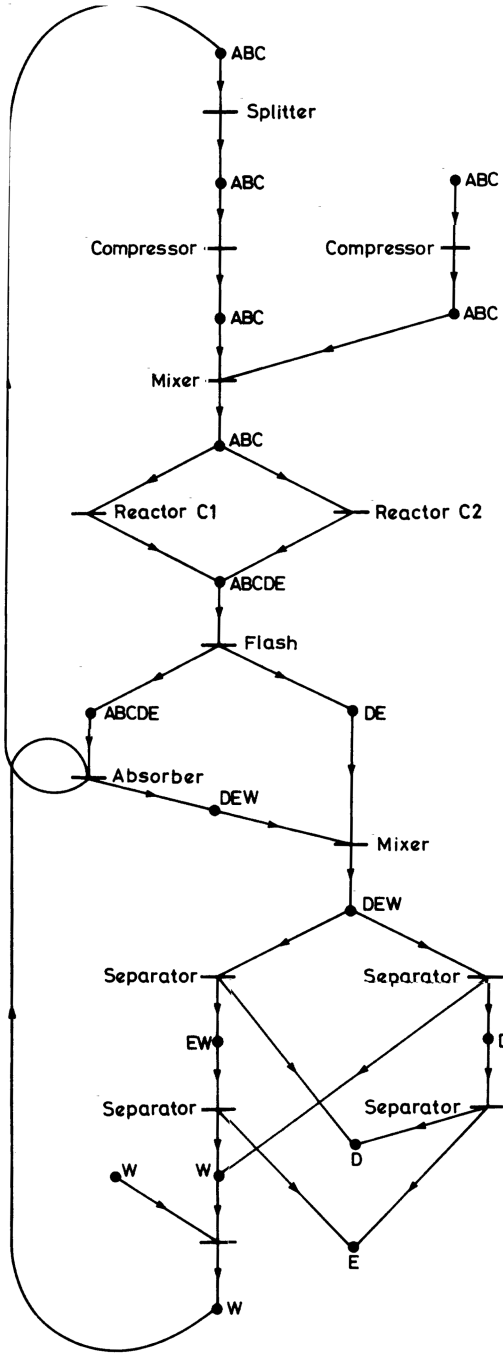

Fig. 16. Maximal structure of the synthesis problem of Grossmann (1985).

Theorem 10 (constructive theorem on the extraction of a maximal structure): *If*

$$\mathscr{P}_1 \subseteq \mathscr{P}_2$$

$$\mathrm{S}(\mathscr{P}_2, \mathscr{R}, \mathscr{O}) \neq \varnothing$$

$$\mu(\mathscr{P}_2, \mathscr{R}, \mathscr{O}) = (m_2, o_2)$$

$$o_1 = \{y_0 \mid y_0 \in o_2 \text{ and } \exists \text{ path } [y_0, y_1] \text{ in } (m_2, o_2) \text{ such that } y_1 \in \mathscr{P}_1\}$$

and

$$m_1 = \bigcup_{(\alpha,\beta)\in o_1} (\alpha \cup \beta)$$

then

$$(m_1, o_1) = \mu(\mathscr{P}_1, \mathscr{R}, \mathscr{O}).$$

Theorem 11 (contractibility of maximal structures): *A maximal structure generating a specific set of products can be extracted from the intersection of two maximal structures, each generating a complementary part of the products contained in the set, that is,*

$$\mu(\mathscr{P}_1 \cap \mathscr{P}_2, \mathscr{R}, \mathscr{O}) \subseteq \mu(\mathscr{P}_1, \mathscr{R}, \mathscr{O}) \cap \mu(\mathscr{P}_2, \mathscr{R}, \mathscr{O}).$$

Examples

By theorem 7, a maximal structure is a solution-structure; however, usually it may not necessarily be the optimal one. For example, the maximal structure of Fig. 13 satisfies all the axioms of solution-structure for synthesis problem $(\mathscr{P}_3, \mathscr{R}_3, \mathscr{O}_3)$; therefore, it is a solution-structure for the problem. Similarly, the maximal structure of Fig. 14 satisfies all the axioms of solution-structure for synthesis problem $(\mathscr{P}_4, \mathscr{R}_4, \mathscr{O}_4)$; it is a solution-structure for the problem.

According to theorem 8, the maximal structures of two synthesis problems, $(\mathscr{P}_1, \mathscr{R}, \mathscr{O})$ and $(\mathscr{P}_2, \mathscr{R}, \mathscr{O})$, can be combined to yield the maximal structure for the larger synthesis problem, $(\mathscr{P}_1 \cup \mathscr{P}_2, \mathscr{R}, \mathscr{O})$. For example, the union of the maximal structure of synthesis problem $(\{A, B\}, \mathscr{R}_4, \mathscr{O}_4)$, depicted in Fig. 17, and that of problem $(\{C\}, \mathscr{R}_4, \mathscr{O}_4)$, depicted in Fig. 18, yield the structure given in Fig. 14, which is the maximal struc-

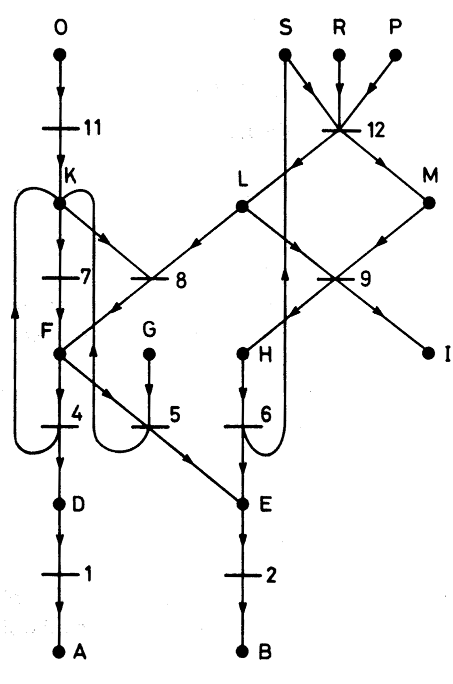

Fig. 17. Maximal structure of synthesis problem $(\{A, B\}, \mathscr{R}_4, \mathscr{O}_4)$ (I: by-product).

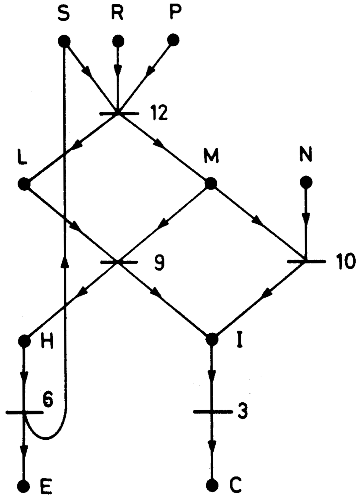

Fig. 18. Maximal structure of synthesis problem ($\{C\}$, $\mathscr{R}_4$, $\mathscr{O}_4$) (E: by-product).

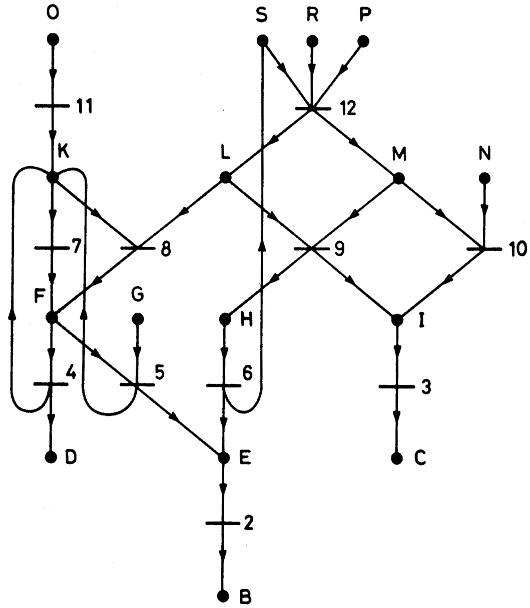

Fig.19. Maximal structure of synthesis problem ($\{B, C\}$, $\mathscr{R}_4$, $\mathscr{O}_4$) (D: by-product).

ture for problem ($\mathscr{P}_4$, $\mathscr{R}_4$, $\mathscr{O}_4$). Note that

$$\mathscr{P}_4 = \{A, B, C\} = \{A, B\} \cup \{C\}.$$

Also note that material I in Fig. 17 and material E in Fig. 18 are not defined as products in synthesis problem ($\mathscr{P}_4$, $\mathscr{R}_4$, $\mathscr{O}_4$). They are by-products generated by the synthesized processes. A by-product is a secondary product obtained in addition to the primary products to be generated by the process being synthesized or designed.

Theorem 9 states that the maximal structure of a smaller synthesis problem, ($\mathscr{P}_1$, $\mathscr{R}$, $\mathscr{O}$), can be abridged from the maximal structure of a larger problem, ($\mathscr{P}_2$, $\mathscr{R}$, $\mathscr{O}$), where

$$\mathscr{P}_1 \subseteq \mathscr{P}_2.$$

For example, the maximal structure of synthesis problem ($\{A, B\}$, $\mathscr{R}_4$, $\mathscr{O}_4$) in Fig. 17 is contained in the maximal structure of synthesis problem ($\mathscr{P}_4$, $\mathscr{R}_4$, $\mathscr{O}_4$) in Fig. 14 with

$$\{A, B\} \subset \mathscr{P}_4.$$

Theorem 10 essentially elaborates the mathematical basis for constructing an algorithm to determine $\mu(\mathscr{P}_1, \mathscr{R}, \mathscr{O})$ in theorem 9.

Theorem 11 asserts that a subgraph of the common portion of the maximal structures of two synthesis problems, ($\mathscr{P}_1$, $\mathscr{R}$, $\mathscr{O}$) and ($\mathscr{P}_2$, $\mathscr{R}$, $\mathscr{O}$), can be extracted to yield a structure which is contained in the maximal structure for the smaller synthesis problem, ($\mathscr{P}_1 \cap \mathscr{P}_2$, $\mathscr{R}$, $\mathscr{O}$). For example, the intersection of the maximal structure of synthesis problem ($\{A, B\}$, $\mathscr{R}_4$, $\mathscr{O}_4$), depicted in Fig. 17, and that of problem ($\{B, C\}$, $\mathscr{R}_4$, $\mathscr{O}_4$), depicted in Fig. 19, yields the structure given in Fig. 20, which is the maximal structure for problem ($\{B\}$, $\mathscr{R}_4$, $\mathscr{O}_4$). Clearly,

$$\mu(\{B\}, \mathscr{R}_4, \mathscr{O}_4) \subseteq \mu(\{A, B\}, \mathscr{R}_4, \mathscr{O}_4) \cap \mu(\{B, C\}, \mathscr{R}_4, \mathscr{O}_4).$$

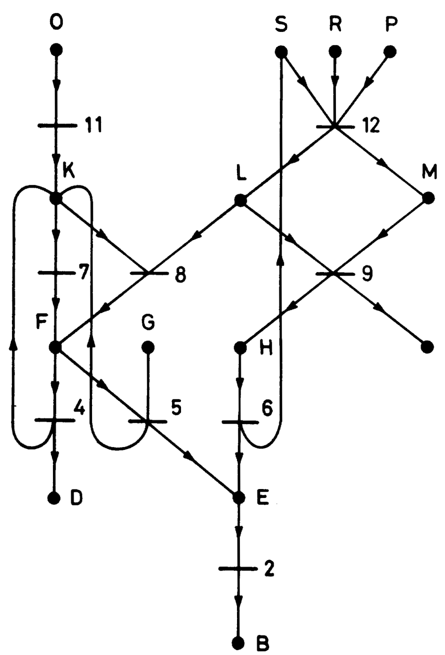

Fig. 20. Maximal structure of synthesis problem ($\{B\}$, $\mathscr{R}_4$, $\mathscr{O}_4$) (D, I: by-products).

Note that material D in Fig. 19 and materials D and I in Fig. 20 are by-products.

CONCLUDING REMARKS

An innovative graph-theoretic approach to process synthesis has been introduced. The heart of the approach is a special directed bipartite graph known as a process graph, or P-graph in brief. An axiom system has been developed to render effective process synthesis possible through manipulation of feasible process structures represented as P-graphs. Such manipulation is governed by a set of theorems derived from this axiom system; the validity of each of these theorems has been proved by exploiting the unique features and fundamental properties of process structures in synthesis. The theorems form the basis for developing additional theorems and for deriving effective algorithms whose validity can also be assured. In principle, the present approach is applicable to the synthesis of any process systems with minimum generation of waste; it is also applicable to the synthesis of systems to be constructed in stages.

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NOTATION

$\mathscr{A}$	set of arcs of a P-graph
d^-	indegree of a vertex
iff, $\Leftrightarrow$	if and only if
(m, o)	P-graph
m, $\mathscr{M}$	set of materials
o, $\mathscr{O}$	set of operating units
$\mathscr{P}$	set of products
$(\mathscr{P}, \mathscr{R}, \mathscr{O})$	synthesis problem defined by the specific sets of products ($\mathscr{P}$), raw materials ($\mathscr{R}$), and operating units ($\mathscr{O}$)
$\mathscr{R}$	set of raw materials
$S(\mathscr{P}, \mathscr{R}, \mathscr{O})$	set of solution-structures for synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$
$\mathscr{V}$	set of vertices of a P-graph
$[y_i, y_j]$	path in a P-graph

Greek letters

$\mu(\mathscr{P}, \mathscr{R}, \mathscr{O})$	maximal structure for synthesis problem $(\mathscr{P}, \mathscr{R}, \mathscr{O})$
σ	solution-structure

Mathematical symbols

$\emptyset$	empty set
$\wp$	power set
$\forall$	for any
$\exists$	there exists
$\Rightarrow$	imply
$\square$	end of proof
$\times$	Cartesian product
$\{\ \}$	set
$\lvert\ \rvert$	cardinality of a set
$\setminus$	set difference
$(\subset)\ \subseteq$	(proper) subset or subgraph
$(\supset)\ \supseteq$	(proper) superset or supergraph
$\in$	element
$\notin$	not an element
$\cap$	intersection of sets or graphs
$\cup$	union of sets or graphs
$\bigcup_{i \in I} \chi_i$	union of all the elements of sets χ_i $(i \in I)$

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APPENDIX A: FORMAL DEFINITIONS

Let $\mathscr{M}$ be a finite nonempty set.

Synthesis problem

A *synthesis problem* is defined by triplet $(\mathscr{P}, \mathscr{R}, \mathscr{O})$, where

$$\mathscr{P}(\subset \mathscr{M})$$

is a set of final products,

$$\mathscr{R}(\subset \mathscr{M}, \mathscr{P} \cap \mathscr{R} = \emptyset)$$

is a set of raw materials, and

$$\mathscr{O} \subseteq (\wp(\mathscr{M}) \times \wp(\mathscr{M}))$$

is a set of operating units.

Process graph (P-graph)
Let m be a finite set and

$$o \subseteq \wp(m) \times \wp(m).$$

A *P-graph* is defined by pair (m, o) where *vertices* of the graph are the elements of

$$m \cup o$$

and *arcs* of the graph are the elements of

$$\mathcal{A} = \mathcal{A}_1 \cup \mathcal{A}_2$$

with

$$\mathcal{A}_1 = \{(x, y) | y = (\alpha, \beta) \in o \text{ and } x \in \alpha\}$$

and

$$\mathcal{A}_2 = \{(y, x) | y = (\alpha, \beta) \in o \text{ and } x \in \beta\}.$$

Moreover, $[y_0, y_n]$ is a *path* in P-graph (m, o) if

$$(y_{i-1}, y_i) \in \mathcal{A}, \qquad i = 1, 2, 3, \ldots, n.$$

The *union* and *intersection* of two P-graphs, (m_1, o_1) and (m_2, o_2), are defined, respectively, by

$$(m_1, o_1) \cup (m_2, o_2) = (m_1 \cup m_2, o_1 \cup o_2)$$

and

$$(m_1, o_1) \cap (m_2, o_2) = (m_1 \cap m_2, o_1 \cap o_2).$$

Graph (m_1, o_1) is a *subgraph* of graph (m_2, o_2), i.e.

$$(m_1, o_1) \subseteq (m_2, o_2)$$

if

$$(m_1 \subseteq m_2) \quad \text{and} \quad (o_1 \subseteq o_2).$$

Let (m, o) be a P-graph. The *indegree* of a vertex, $\chi (\in m)$, is defined by

$$d^-(\chi) = |o'|$$

where

$$o' = \{(\alpha, \beta) | (\alpha, \beta) \in o \text{ and } \chi \in \beta\}.$$

Structure of a system
Let $m \subseteq \mathcal{M}$, $o \subseteq \mathcal{O}$ and $o \subseteq \wp(m) \times \wp(m)$. The *structure* of this system is defined by P-graph (m, o).

Solution-structure
P-graph (m, o) is a *solution-structure* of synthesis problem $(\mathcal{P}, \mathcal{R}, \mathcal{O})$ if it satisfies the following axioms:

(S1) $\mathcal{P} \subseteq m$
(S2) $\forall \chi \in m, d^-(\chi) = 0$ iff $\chi \in \mathcal{R}$
(S3) $o \subseteq \mathcal{O}$
(S4) $\forall y_0 \in o, \exists$ path $[y_0, y_1]$, where $y_1 \in \mathcal{P}$
(S5) $\forall \chi \in m, \exists (\alpha, \beta) \in o$ such that $\chi \in (\alpha \cup \beta)$.

The set of solution-structures of synthesis problem $(\mathcal{P}, \mathcal{R}, \mathcal{O})$ will be denoted by $S(\mathcal{P}, \mathcal{R}, \mathcal{O})$.

Maximal structure
The P-graph, $\mu(\mathcal{P}, \mathcal{R}, \mathcal{O})$, is defined to be the *maximal structure* of synthesis problem $(\mathcal{P}, \mathcal{R}, \mathcal{O})$ by the following equation.

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

APPENDIX B: PROOF OF THEOREMS

The proofs resort to the following four propositions.

Proposition 1: *Let P-graph (m, o) be given. Then,*

$$\forall \chi \in m, \exists (\alpha, \beta) \in o$$

such that

$$\chi \in (\alpha \cup \beta)$$

if and only if

$$m \subseteq \bigcup_{(\alpha, \beta) \in o} (\alpha \cup \beta). \tag{B1}$$

Hence, axiom (S5) can be replaced by the expression above.

Lemma: *Let (m, o) be a P-graph and let expression (B1) hold. 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

$$m = \bigcup_{(\alpha, \beta) \in \wp(m) \times \wp(m)} (\alpha \cup \beta) \supseteq \bigcup_{(\alpha, \beta) \in o} (\alpha \cup \beta). \tag{B2}$$

Expressions (B1) and (B2) lead to the lemma. □

Corollary: *If P-graph (m, o) satisfies axiom (S5), then it can be defined by the set o.*

Propositon 2: *Let*

$$\sigma = (m, o) \in S(\mathcal{P}, \mathcal{R}, \mathcal{O})$$
$$\mathcal{P}' \subset \mathcal{P}$$

$$o' = \{y_0 | y_0 \in o \text{ and } \exists \text{ path } [y_0, y_1] \text{ in } \sigma \text{ such that } y_1 \in \mathcal{P}'\}$$

$$m' = \bigcup_{(\alpha, \beta) \in o'} (\alpha \cup \beta)$$

and

$$\sigma' = (m', o').$$

Then

$$\forall \chi \in m', d^-_\sigma(\chi) = 0 \Leftrightarrow d^-_{\sigma'}(\chi) = 0.$$

Proof: Let

$$m'' = \bigcup_{\substack{(\alpha, \beta) \in o' \\ \chi \in m''}} \alpha \supseteq \{\chi | \chi \in m' \text{ and } d^-(\chi) = 0\}$$

and

$$o'' = \{(\alpha, \beta) | (\alpha, \beta) \in o \text{ and } \chi \in \beta\}.$$

Then

and

$$o'' \subseteq o'$$

and thus,

$$d^-_\sigma(\chi) = |o''| = d^-_{\sigma'}(\chi).$$ □

Propositon 3: *Let*

$$\sigma = (m, o) \in S(\mathcal{P}, \mathcal{R}, \mathcal{O})$$
$$\mathcal{P}' \subset \mathcal{P}$$

$$o' = \{y_0 | y_0 \in o \text{ and } \exists \text{ path } [y_0, y_1] \text{ in } \sigma \text{ such that } y_1 \in \mathcal{P}'\}$$

$$m = \bigcup_{(\alpha, \beta) \in o'} (\alpha \cup \beta)$$

and

$$\sigma' = (m', o').$$