A State Space Modeling and Evolutionary Programming Approach to Automatic Synthesis of Chemical Processes

Soo Hyoung Choi*, Bomsock Lee**, and Chang-Bock Chung***

* Division of Environmental and Chemical Engineering, Chonbuk National University, Jeonju, Korea
(Tel : +82-63-270-2432; E-mail: soochoi@chonbuk.ac.kr)
** School of Environment and Applied Chemistry, Kyung Hee University, Yongjin, Korea
(Tel : +82-31-201-2598; E-mail: bslee@khu.ac.kr)
*** Faculty of Applied Chemical Engineering, Chonnam National University, Gwangju, Korea
(Tel : +82-62-530-1884; E-mail: chungcb@chonnam.ac.kr)

Abstract: The objective of this study is to investigate the possibility of chemical process synthesis purely based on mathematical programming when given an objective, feed conditions, product specifications, and model equations for available process units. A method based on a state space approach is proposed, and applied to an example problem with a reactor, a heat exchanger, and a separator. The results indicate that a computer can automatically synthesize an optimal process without any heuristics or expertise in process design provided that global optimization techniques are improved to be suitable for large problems.

Keywords: process synthesis, state space, superstructure, mathematical programming, global optimization

1. INTRODUCTION

Chemical process design is generally performed in two steps. First, a flowsheet is generated by structural optimization, which is usually done by heuristics or expertise in chemical process design. Second, the values of the design variables in the flowsheet are determined by parametric optimization, which is usually done by mathematical programming. However, structural optimization can also be based on mathematical programming. For example, Bagajewicz et al. [1] presented a state space approach to mass/heat exchanger network synthesis, and Schweiger and Floudas [2] presented a superstructure method for reactor network synthesis. Their work was focused on synthesis of networks composed of the same kind of units such as reactors, heat exchangers, or separators. Their methods are generalized and extended in this work, and a method is proposed for synthesis of a new chemical process that satisfies a given objective and constraints. Mathematical formulation of a process synthesis problem using the proposed method results in a nonconvex nonlinear program. Solution to this problem automatically removes unnecessary streams and units, and thus the structure is optimized. Furthermore, at the same time, the design and state variables such as a reactor volume and a stream temperature are also optimized. As a result, the structure, size, and operating conditions are simultaneously optimized.

2. PROPOSED METHOD

2.1 Modeling

The method developed in this study is based on the state space approach using the superstructure operator, which is the most general method for representing a flowsheet that contains all the possible structures of a process. This method represents the superstructure of a process to be synthesized as shown in Fig. 1, where the block of process units contains all the available units, and the block of pipe network contains all the possible connection streams. This is to be optimized by solution of a mathematical programming problem which consists of the mass and energy balance equations for the mixers and splitters on the boundaries of the block of pipe network, model equations for the units in the block of process units, constraints on materials and products, and the objective function that represents the goal of the process synthesis.

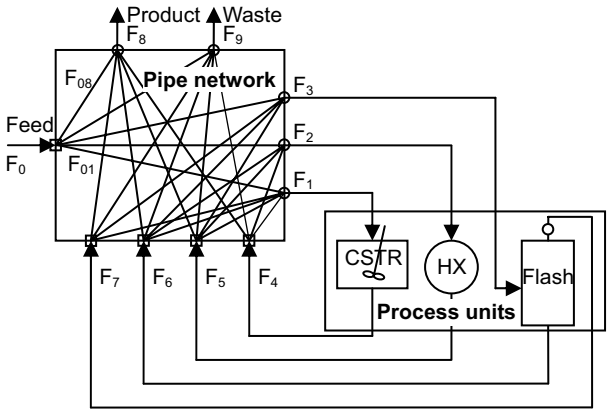

Fig. 1 Process superstructure by state space approach.

Let us represent each splitter in the pipe network as $i \in I$ and mixer as $j \in J$. The input stream to splitter i is represented as F_i, the output stream from mixer j as F_j, and the stream from splitter i to mixer j as F_{ij}. These stream symbols will also be used to represent their molar flowrates. Each component in the stream is represented as $k \in K$, the component molar flowrate of each stream as F_{ik} and F_{jk} outside the network, and as F_{ijk} inside the network. The temperature and the pressure of stream F_i are represented as T_i and P_i respectively, and the fraction of F_i that is connected to F_j as $a_{ij} \in [0, 1]$. Each available unit is represented as $l \in L$, and their parameters as $\mathbf{w}_l$. Let us simply represent the variables as $\mathbf{F} = \{F_{ik} \mid i \in I \cup J, k \in K\}$, $\mathbf{T} = \{T_i \mid i \in I \cup J\}$, $\mathbf{P} = \{P_i \mid i \in I \cup J\}$, $\mathbf{A} = \{a_{ij} \mid i \in I, j \in J\}$, and $\mathbf{W} = \{\mathbf{w}_l \mid l \in L\}$. Then the problem is as follows.

$$\min \quad f(\mathbf{F},\mathbf{T},\mathbf{P},\mathbf{A},\mathbf{W})$$

subject to

Pipe network:

$$F_{ijk} = a_{ij} F_{ik}, \quad i \in I, j \in J, k \in K$$

$$\sum_{j \in J} a_{ij} = 1, \quad i \in I$$

$$\sum_{i \in I} F_{ijk} = F_{jk}, \quad j \in J, k \in K$$

$$\sum_{i \in I} F_{ij} H(F_{ik}, T_i, P_i) = F_j H(F_{jk}, T_j, P_j), \quad j \in J$$

Process units:

$$\mathbf{h}_l(\mathbf{F},\mathbf{T},\mathbf{P},\mathbf{w}_l) = \mathbf{0}, \quad l \in L$$

Other constraints:

$$\mathbf{g}(\mathbf{F},\mathbf{T},\mathbf{P},\mathbf{W}) \le \mathbf{0}$$

$$\mathbf{F},\mathbf{T},\mathbf{P},\mathbf{A} \ge \mathbf{0}$$

where function H represents the molar enthalpy of the corresponding stream. The objective function f is eventually to be set in such a way that the profit is maximized. However, at the beginning stage of the design, a simpler objective function can be used, for example, to maximize the yield of the product, or to minimize the waste. The following objective function is proposed in this paper in order to find the simplest process that discharges the minimum waste.

$$f = c \sum_{k \in K} F_{nk} - \sum_{i \in I} \sum_{j \in J} a_{ij}^{\ 2}$$

where coefficient c is a sufficiently large positive constant. Therefore, the first term is much larger than the second, and thus the waste flowrate F_n is minimized first. The second term tries to increase one of the values of a_{ij}'s for each splitter i to its upper bound value, 1. This is to remove unnecessarily split streams when the first term is constant, and thus simplify the process. The variables that have been used so far are all continuous. Integer variables could be introduced, for example, in order to take into account whether a unit is used or not, but not in this work.

2.2 Optimization

The problem formulated above is a nonconvex nonlinear program, and in most cases, has multiple local optima. Therefore, it is necessary to find the global optimum among them. However, global optimization of a nonconvex program is an NP-hard problem for which the worst case computation time exponentially increases with the size of the problem. Moreover, many nonlinear equality constraints are involved because of the model equations for the units, and thus this problem is a particularly tough one among the NP-hard problems. Therefore, in order to deal with large problems, deterministic methods are avoided, which guarantee the global optimality of the obtained solution, and stochastic approach is adopted, which does not suffer from the NP-hard property [3].

A stochastic global optimization algorithm proposed in this paper is as follows.

Step 0: Initialization
1) Define constraint $f(\mathbf{x}) \le f^* - \varepsilon$ where $\varepsilon > 0$.
2) Set $f^* = +\infty$.

Step 1: Starting point generation
1) Generate random points in the search space.
2) Select the least infeasible one as starting point.

Step 2: Infeasibility minimization
1) Find a feasible point.
2) If no feasible point is found, go to step 1 or stop.

Step 3: Objective function minimization
1) Find a local minimum f^*.
2) If a better solution is wanted, go to step 1.

The constraint defined in step 0 forces the optimizer to find a lower point than the currently known local minimum. In step 1, a stochastic global optimization method can be used such as simulated annealing, genetic algorithm, and Tabu search. In steps 2 and 3, a deterministic local optimizer is used which is based on sequential quadratic programming or generalized reduced gradient method. The proposed algorithm continually improves the process, and thus can be viewed as an evolution program, especially when genetic algorithm is adopted in step 1. In this paper, no specific algorithm is applied to step 1, and the procedure is manually implemented in search of a better local solution.

3. CASE STUDY

A process is to be synthesized that produces B from A using a reaction A (feed) → B (product) → C (byproduct). The physical properties and reaction data are listed in Tables 1 and 2 [4]. The units available for this process are an isothermal CSTR (continuous stirred tank reactor), a heat exchanger, and an adiabatic flash. The reactor can contain a volume of 10 ft^3 at maximum, and its maximum temperature is 900 °R. The flash is to be operated at pressure of 14.7 psia, and the maximum flowrate into it is 300 lb-moles/hr. The feed stream is composed of A only, the flowrate is 100 lb-moles/hr, and the temperature is 537 °R. The objective is to design a process that produces a product in which the mole fraction of B is at least 95%, minimizing the waste flowrate.

Table 1 Physical property data.

Component (i)	A	B	C
Density ρ_i, lb-mole/ft^3	0.667	0.667	0.667
Heat of vaporization λ_i, Btu/lb-mole	17,580	16,150	18,060
Heat capacity C_i, Btu/(lb-mole °R)	39.60	40.48	45.76
Vapor pressure correlation $\ln P_i^s$ (psia) = $A_i - B_i / T$			
A_i	15.84	14.88	16.46
B_i, °R	10,141.2	7,689.2	10,841.1

Table 2 Reaction data.

No. (i)	1	2
Reaction	A → B	B → C
Rate	$r_1 = k_1$ [A]	$r_2 = k_2$ [B]^2
Rate constant $k_i = a_i \exp(-E_i / (RT))$		
a_i	0.1269×10^{12} hr^{-1}	0.3077×10^7 ft^3/(lb-mole hr)
E_i / R	17,900 °R	11,910 °R

The superstructure based on the state space approach which contains all processes that can be constructed using the given units is as shown in Fig. 1. Let us assume that all the streams represented by arrows in the figure are liquids. Therefore, the gas stream from the top of the flash is to be immediately condensed to a saturated liquid. Furthermore, the enthalpies are assumed to be functions of temperatures only, and the pressures of the streams are not considered. Let us now define the following sets that represent the splitters, the mixers, and the components respectively.

$$I = \{0, 4, 5, 6, 7\}$$
$$J = \{1, 2, 3, 8, 9\}$$
$$K = \{\mathrm{A}, \mathrm{B}, \mathrm{C}\}$$

Then the problem is formulated as follows.

$$\min f = c\sum_{k\in K} F_{9k} - \sum_{i\in I}\sum_{j\in J} a_{ij}^{\ 2}$$

subject to

Splitters:

$$F_{ijk} = a_{ij}F_{ik}, \quad i \in I, j \in J, k \in K$$
$$\sum_{j\in J} a_{ij} = 1, \quad i \in I$$

Mixers:

$$\sum_{i\in I} F_{ijk} = F_{jk}, \quad j \in J, k \in K$$
$$\sum_{i\in I} T_i \sum_{k\in K} C_k F_{ijk} = T_j \sum_{k\in K} C_k F_{jk}, \quad j \in J$$

Reactor:

$$R_1 = a_1 e^{-\frac{E_1}{RT_R}} \frac{\rho_R F_{4\mathrm{A}}}{F_{4\mathrm{A}} + F_{4\mathrm{B}} + F_{4\mathrm{C}}} V_R$$
$$R_2 = a_2 e^{-\frac{E_2}{RT_R}} \left(\frac{\rho_R F_{4\mathrm{B}}}{F_{4\mathrm{A}} + F_{4\mathrm{B}} + F_{4\mathrm{C}}} \right)^2 V_R$$
$$F_{1\mathrm{A}} - F_{4\mathrm{A}} - R_1 = 0$$
$$F_{1\mathrm{B}} - F_{4\mathrm{B}} + R_1 - R_2 = 0$$
$$F_{1\mathrm{C}} - F_{4\mathrm{C}} + R_2 = 0$$
$$T_4 = T_R$$

Heat exchanger:

$$F_{2k} = F_{5k}, \quad k \in K$$
$$(T_2 - T_5)\sum_{k\in K} C_k F_{2k} + Q_H = 0$$

Flash:

$$F_{3k} = F_{6k} + F_{7k}, \quad k \in K$$
$$(T_3 - T_F)\sum_{k\in K} C_k F_{3k} - \sum_{k\in K} \lambda_k F_{7k} = 0$$
$$\frac{F_{7k}}{\sum_{i\in K} F_{7i}} P_F = \frac{F_{6k}}{\sum_{i\in K} F_{6i}} e^{A_k - \frac{B_k}{T_F}}, \quad k \in K$$
$$T_6 = T_F; \quad T_7 = T_F$$

Specifications:

$$\frac{F_{8B}}{F_{8A} + F_{8B} + F_{8C}} \geq 0.95$$
$$V_R \leq 10, \quad T_R \leq 900$$
$$F_{3A} + F_{3B} + F_{3C} \leq 300$$

The coefficient in the objective function is set to $c = 100$. The variable Q_H used in one of the constraints represents the heat duty of the heat exchanger. If its value is positive, it means to heat, and if negative, to cool. This problem has 119 equality constraints and 4 inequality constraints. If we substitute values for all known variables such as $F_{0\mathrm{A}} = 100$, $F_{0\mathrm{B}} = F_{0\mathrm{C}} = 0$, $T_0 = 537$, $\rho_R = 0.667$, and $P_F = 14.7$, there remain 142 unknowns. In order to improve the solution efficiency and robustness, the lower and upper limits were provided for all these variables. For example, the temperatures were assumed to be above 500 °R. A local optimizer LINGO was used to solve this problem.

4. RESULTS AND DISCUSSION

A local solution was found by optimization, which is shown in Tables 3 and 4. This solution corresponds to the flowsheet in Fig. 2. It is shown that we can design a process that meets the product specification without using the heat exchanger. However, stream F_{73} was used, which recycles a portion of the top stream from the flash back into the flash. Without this, the flowsheet would have the same structure as in the source of data for this example problem [4]. In this case, much more waste should be discharged in order to achieve the 95 mol% purity of the product. In order to verify it, a constraint $a_{73} = 0$ was added, and the problem was solved again. As a result, the waste flowrate increased to $F_9 = F_{69} = 16.782$ lb-moles/hr. The use of stream F_{73} is not a common application to a flash, but in principle, it is similar to increasing the reflux ratio of a distillation column in order to increase the purity of the product.

Table 3 Network data of local solution 1.

i	a_{i1}	a_{i2}	a_{i3}	a_{i8}	a_{i9}
0	1	0	0	0	0
4	0	0	1	0	0
5	0	0	0	1	0
6	0.9627	0	0	0	0.0373
7	0	0	0.3599	0.6401	0

Table 4 Stream data of local solution 1.

i	$F_{i\mathrm{A}}$	$F_{i\mathrm{B}}$	$F_{i\mathrm{C}}$	T_i
1	131.151	73.347	42.485	615.35
2	0	0	0	500.00
3	36.265	216.142	47.594	859.25
4	34.859	165.776	46.348	900.00
5	0	0	0	500.00
6	32.359	76.192	44.133	665.83
7	3.905	139.950	3.461	665.83
8	2.500	89.585	2.215	665.83
9	1.208	2.845	1.648	665.83

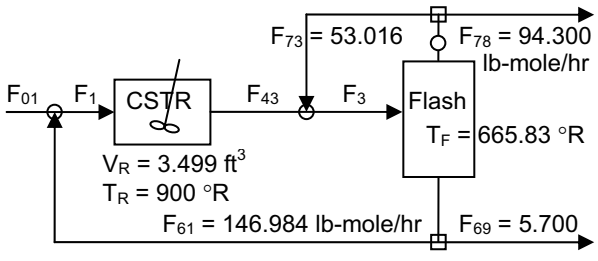

Fig. 2 Process flowsheet by local solution 1.

Another local solution was found, which is shown in Tables 5 and 6. This solution corresponds to the flowsheet in Fig. 3. The product purity is the same as before, but the waste was reduced. However, unlike the previous design, it requires a heat exchanger and cooling water in addition. The value of the objective function at this point is $f = 355.81$. The search was continued, and a better local solution was found at $f = 355.42$. This solution corresponds to Table 5 with $a_{42} = 1$ and $a_{43} = 0$ in it, and Table 6 with the F_2 and F_5 data replaced by the F_4 and F_3 data respectively. At this time, the first term in the objective function was unchanged, but the second term was improved. As a result, the bypass stream F_{43} is removed, and thus the process is simplified.

Table 5 Network data of local solution 2.

i	a_{i1}	a_{i2}	a_{i3}	a_{i8}	a_{i9}
0	1	0	0	0	0
4	0	0.2706	0.7294	0	0
5	0	0	1	0	0
6	0.9823	0	0	0	0.0177
7	0	0	0	1	0

Table 6 Stream data of local solution 2.

i	$F_{i\mathrm{A}}$	$F_{i\mathrm{B}}$	$F_{i\mathrm{C}}$	T_i
1	141.716	99.668	58.617	624.50
2	12.171	52.228	16.767	900.00
3	44.987	193.040	61.973	791.78
4	44.987	193.040	61.973	900.00
5	12.171	52.228	16.767	500.00
6	42.467	101.464	59.673	665.91
7	2.519	91.576	2.300	665.91
8	2.519	91.576	2.300	665.91
9	0.752	1.796	1.056	665.91

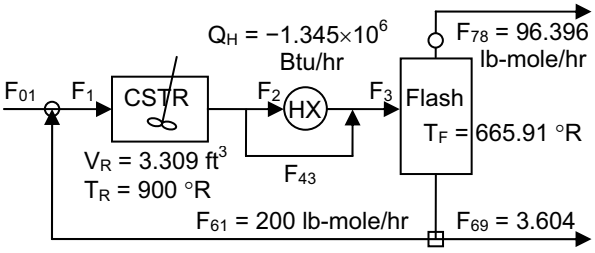

Fig. 3 Process flowsheet by local solution 2.

Note that no heuristic rules or expertise in process design was used during the solution of this example problem. However, a product reflux technique automatically appeared as shown in Fig. 2, and the final result, which corresponds to Fig. 3 without stream F_{43} , shows the structure of a typical chemical process such as Williams-Otto plant. This process is considered to be the global optimum, but not guaranteed unless solved by a deterministic method.

5. CONCLUSION

The state space and superstructure methods used to be applied to synthesis of networks of the same kind of specific units such as reactors, heat exchangers, or separators. A method has been proposed in this paper, which applies the state space modeling technique to a general process synthesis problem. A stochastic global optimization algorithm suitable for this type of problem is also proposed, which continually improves local optima. A case study showed us local solutions evolving to a flowsheet that represents a process with a reasonable structure and feasible operating conditions. This result indicates the possibility that when the objective and conditions for materials and products are given, the computer can automatically synthesize an optimal process using the model equations for the available units. Therefore, the proposed method is expected to be able to contribute to synthesis of a new process for new materials and products, provided that global optimization techniques are improved so that larger problems can be solved.

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