A Multilevel MINLP Methodology for the Approximate Stochastic Synthesis of Flexible Chemical Processes

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This work presents a methodology for the optimal design and MINLP synthesis of flexible chemical processes with known probability distributions of uncertain parameters. This methodology comprises synthesis at 1) non-flexible nominal level, 2) flexible nominal level, and 3) approximate stochastic flexible level. First level is concerned with modeling challenges of large process flow sheets, sensitivity analyses and studies of uncertain parameters. This level usually results in non-flexible design. Second and third levels take into account flexibility requirements by adding critical points into the model. Both flexible levels rely on considerable reduction of discrete points. Lower levels provide good starting structures for higher levels, therefore, the computational effort is reduced and larger problems with many uncertain parameters, e.g. 10 to 100, can be solved. The use of this methodology is illustrated by the synthesis of a flexible heat-integrated methanol process flow sheet.

Keywords: process design, synthesis, flexible, stochastic, uncertain, MINLP

Chemical engineers have to cope with the presence of uncertainty in almost every activity that concerns the decision making process, e.g. from the planning, scheduling, design and synthesis problems to the financial, strategic and global supply chain and enterprise-wide network optimization problems [1-2].

Several approaches have been developed for various complex problems comprising uncertainty, from the classical recourse-based models to fuzzy programming, with a combination of global optimization algorithms [3]. Despite the significant progress in the field, the design and, in particular, the synthesis of large flexible process flow sheets with a significant number of uncertain parameters is still a challenging problem. The main reason is that such problems are usually solved by the discretization of an infinite uncertain space, which may cause an enormous increase in a problem's size. Several authors have proposed various approaches for facilitating the process synthesis under uncertainty [4-6]. However, a step forward should be taken in order to relate flexible synthesis to real-size applications.

Majority of chemical processes that engineers would like to synthesize or optimize in the practice have much larger numbers of uncertain parameters than it would be possible to handle with the exact mathematical approaches. Therefore, several empirical and/or approximate mathematical approaches have been developed, e.g. [7].

Two main problems we are faced when dealing with flexibility of chemical processes are nonoptimality and infeasibility. E.g., if the sizes of process equipment are increased by some empirical oversizing factors, in order to attain requested flexibility, designs obtained would be most probably near optimal or even nonoptimal. Secondly, if we optimize design at nominal conditions, the solution obtained would be most probably incapable to tolerate variations of a large number of uncertain parameters, which means infeasible solutions for some realizations of uncertain parameters.

The main purpose of this contribution is to develop a robust and reliable strategy for the Mixed Integer Nonlinear Programming (MINLP) synthesis of flexible process flow sheets, which can solve those larger synthesis problems having a considerable number of uncertain parameters. The methodology starts with less accurate (non-flexible) level and gradually proceeds to more accurate flexible synthesis. Lower levels provide good starting structures for higher levels and the computational effort is thus reduced. Flexible levels rely on discretization of uncertain space, however, the number of discretized points is significantly smaller than by more rigorous stochastic methods.

The methodology will be illustrated by the three-steps synthesis of a flexible heat-integrated methanol process flow sheet with 24 uncertain parameters. Similar approach can be applied also to Nonlinear Programming (NLP) design when process flow sheets are considered at fixed topology.

Methodology description

The main idea is to perform the synthesis through several levels, from a less accurate simple level to more accurate, approximate stochastic level, which is computationally more demanding. As the lower levels generate good initial structures for upper levels, the latter needs less iterations and the computational effort can be significantly reduced. In our work, a three-level approach has been developed:

a)MINLP level 1: Deterministic non-flexible synthesis

b)MINLP level 2: Nominal flexible synthesis

c)MINLP level 3: Approximate-stochastic flexible synthesis

MINLP level 1 serves as a basic level, at which mathematical model of flow sheet is developed and uncertain parameters are defined. At the MINLP level 2, the simplified flexible synthesis is performed which gives good starting and flexible structure for the final, third level at which fine tuning of optimal solution is performed by applying the approximate stochastic optimization.

Syntheses at all levels are performed by means of an MINLP algorithm, e.g. Outer Approximation/Equality Relaxation algorithm as shown in figure 1. All three levels are represented in the continuation.

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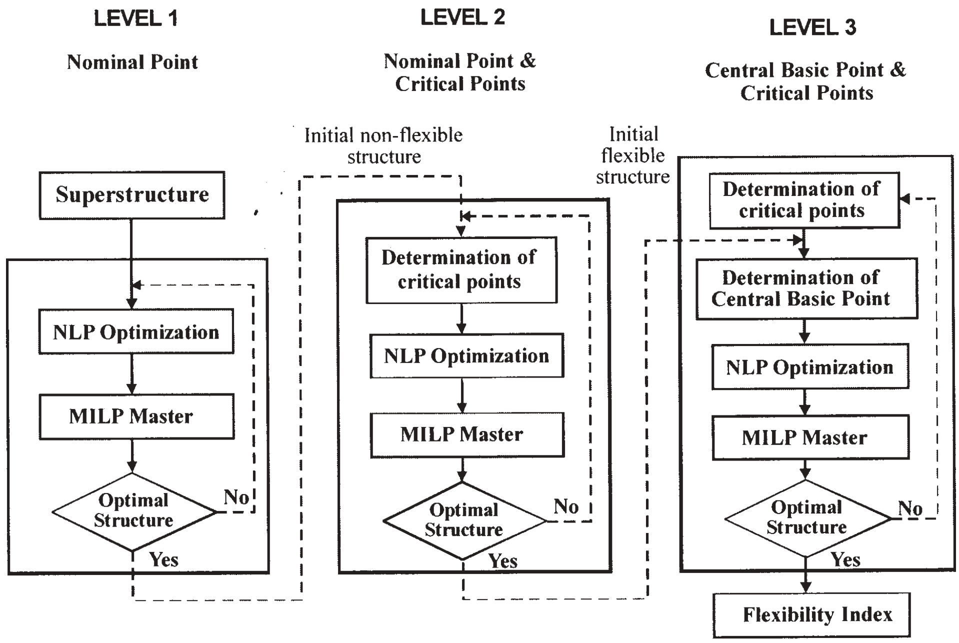

Fig. 1. Three-level strategy for the flexible MINLP synthesis

MINLP level 1 - Deterministic non-flexible synthesis

At the first level, the synthesis is performed at the nominal values of uncertain parameters, θ^{N}, without any consideration of flexibility as shown in the model (P1).

$$
\begin{aligned}
&\min_{y,x,z,d} C(y, x, z, d, \theta^{N}) \\
&\text{s.t.} \\
&h(y, x, z, d, \theta^{N}) = 0 \\
&g(y, x, z, d, \theta^{N}) \le 0 \qquad \text{(P1)} \\
&d = g_d(x, z, \theta^{N}) \\
&x, z, d \in R,\; y \in \{0,1\}^{m}
\end{aligned}
$$

In the model (P1), $\boldsymbol{y}$ represents vector of binary variables for the selection of process topology. $\boldsymbol{x}$, $\boldsymbol{z}$ and $\boldsymbol{d}$ are the vectors of the state, control and design variables (sizes of process units), respectively. Vector of uncertain parameters is denoted by q, while superscript N denotes their nominal values. C is the economic objective function, $\boldsymbol{g}$ and $\boldsymbol{h}$ are the vectors of (in)equality constraints and $\boldsymbol{g}_{\mathrm{d}}$ represents the design specifications.

The solution of (P1) is almost always a non-flexible optimal structure which is incapable to handle numerous expected variations of input parameters. Anyway, this step is important for model developer in order to study the optimization behavior of mostly large and nonconvex flow sheet models. This may facilitate convergence of optimization algorithm at the higher, more complicated synthesis levels. Besides, sensitivity analyses may be performed in order to establish the influence of various uncertain parameters on the optimal structures and design as well as on the objective function value. Based on these results, a list of uncertain parameters should be written together with their expected deviations.

MINLP Level 2 – Nominal flexible synthesis

At this level, several critical points are added to the nominal point in order to fulfill flexibility requirements during the synthesis. In the basic mathematical model of Level 1 all constraints and variables (with exception of design variables) are indexed over the set of critical points and nominal point (P2). This increases the size of basic model for $(N_{\mathrm{CP}}+1)$-times, where N_{CP} is the number of critical points. Specifications of design variables ($d=g_{\mathrm{d}}$) are transformed into inequalities ($d \ge g_{\mathrm{d}}$), as sizes of equipment have to suit even the most unpleasant realization of uncertain parameters and can not change during the operation.

The indexed model (P2) is then solved simultaneously at the nominal point, $\theta_{\mathrm{ap}} = \theta^{\mathrm{N}}$, and critical points, θ_c. Nominal point is used for simple approximation of the objective function's expected value, while critical points assure sufficient sizes of process equipment for feasible operation.

$$
\begin{aligned}
&\min_{y,x_{\mathrm{ap}},z_{\mathrm{ap}},d} C(y, x_{\mathrm{ap}}, z_{\mathrm{ap}}, d, \theta_{\mathrm{ap}}) \\
&\text{s.t.} \\
&h(y,x_{\mathrm{ap}},z_{\mathrm{ap}},d,\theta_{\mathrm{ap}}) = 0 \qquad h(y,x_c,z_c,d,\theta_c) = 0 \\
&g(y,x_{\mathrm{ap}},z_{\mathrm{ap}},d,\theta_{\mathrm{ap}}) \le 0 \qquad g(y,x_c,z_c,d,\theta_c) \le 0 \qquad c \in CP \\
&d \ge g_d(x^{\mathrm{ap}},z^{\mathrm{ap}},\theta^{\mathrm{ap}}) \qquad d \ge g_d(x_c,z_c,\theta_c) \\
&x_{\mathrm{ap}}, z_{\mathrm{ap}}, x_c, z_c, d \in R,\; y \in \{0,1\}^{m},\; \theta^{\mathrm{LO}} \le \theta_c \le \theta^{\mathrm{UP}},\; \theta_{\mathrm{ap}} = \theta^{\mathrm{N}}
\end{aligned}
$$

The left group of constraints in (P2) represents the optimization at the nominal point, $\theta_{\mathrm{ap}} = \theta^{\mathrm{N}}$, which also appears in the objective function C. The right group of constraints refers to the critical points, θ_c. These points have to be determined in advance for each flow sheet selected by the optimization algorithm, as will be described in the next subsection.

The solution of (P2) is optimal flexible structure, which is used as a starting structure for the final, third MINLP level described in the section 2.3.

Determination of critical points

Critical points in this work are defined as those combinations of uncertain parameters that require the largest overdesign of process units for given deviations of uncertain parameters. Equipment dimensions have to suit all predefined deviations at minimum cost. This means, that the flexibility index of the optimal flexible solution, as defined in the literature [8], has to be equal or very close to 1. In our recent work [9, 10], we proposed various schemes for identification of critical points, however, it has emerged during this work that simplified noniterative formulation is, for now, the most appropriate for large process flow sheets.

This formulation is mathematically described by a non-linear model (P3) where the binary variables are fixed, y^{fx}, according to the temporarily selected flow sheet structure. Uncertain parameters are transformed into variables that can vary between the selected lower and upper bounds, θ^{LO} and θ^{UP}.

$$\begin{aligned}
&\min_{x,z,d,\theta} C(y^{fx},x,z,d,\theta) - M \cdot d_i \\
&\text{s.t.} \quad h(y^{fx},x,z,d,\theta) = 0 \\
&\qquad g(y^{fx},x,z,d,\theta) \le 0 \qquad\qquad (P3)_i \quad i=1,2,...,n_d \\
&\qquad d = g_d(x,z,\theta) \\
&\qquad \theta^{LO} \le \theta \le \theta^{UP} \\
&\quad x,z,d,\theta \in R,\ y^{fx} \in \{0,1\}^m
\end{aligned}$$

Assume that the number of design variables in particular structure is n_d. Then, NLP problem (P3) is solved for n_d-times by searching for the maximum value of each design variable d_i, at minimum cost. This is achieved by subtracting the design variable multiplied by a large scalar *M*, from the cost function *C*. The result of n_d subproblems are the critical values of uncertain parameters which are then merged into the smallest set of critical points.

MINLP Level 3 –Approximate-stochastic flexible synthesis

At the third level, the mathematical model is very similar to (P2) with only difference that the central basic point [11] is used for approximation of the expected objective function instead of the nominal point, $\theta_{ap} = \theta^{CBP}$. Critical points are included in the model simultaneously as at the level 2. The central basic point accounts for possible deviations in the expected value from the nominal point. It is determined through one-dimensional Gaussian integration as described extensively in our previous work [11].

In order to summarize the procedure briefly, it should be emphasized that coordinates of this point are determined by one-dimensional stochastic integration of each uncertain parameter over its Gaussian quadrature points. In this integration the remaining uncertain parameters are held at their nominal values while the critical points are included to assure flexibility. Objective values obtained at five Gaussian points are fitted into the curve which correlates values of particular uncertain parameter with the objective function values. The basic coordinate is then determined from this curve as the value of uncertain parameter at which the optimal objective function is equal to the expected objective function determined during one-dimensional integration. The basic coordinates of all uncertain parameters constitute a vector of central basic point which is used for the approximation of the expected objective function.

Synthesis of flexible heat-integrated methanol process

The proposed methodology was applied for the synthesis of a flexible heat-integrated methanol process (Fig. 2) where methanol is produced from hydrogen and carbon oxide. This example was taken from the literature 12] and the prices were updated. This flow sheet is medium-sized with 32 streams, 4 hot streams in coolers (C1-C4) and 2 cold process streams in heaters (H1, H2). Eight binary variables were used for selection between two feed streams, between one- or two-stage compression of the feed stream, two reactors, and one- or two-stage compression of the recycle stream. Additional 38 binary variables were assigned for the selection of heat matches between process streams, as well as between process streams and utilities in the four-stage MINLP heat-integration superstructure [13].

24 uncertain parameters were defined with nominal values and deviations: annual production, temperatures, pressures, compositions and the prices of the feed streams, product, electricity, steam and cooling water, heat transfer coefficients, conversion parameters in the reactors and efficiencies of the compressors.

MINLP level 1 - Deterministic non-flexible methanol synthesis

Deterministic synthesis at the nominal values of uncertain parameters with no flexibility consideration yielded a solution with a profit of 37.37 MUSD/yr. The optimal structure (Fig. 3) was comprised of more expensive feed stream (FEED-2), double-stage feed compression, cheaper reactor with lower conversion (RCT-1), and one-stage recycle compression. This structure is a threshold problem with two process heat exchangers (HE1, HE2), two coolers (C1, C3), and no heaters. Heat integration between the streams is shown with dashed lines in figure 3. It was determined that even small deviations in the uncertain parameters from the nominal values result in infeasible solutions, which indicates non-flexible structure.

MINLP level 2 - Nominal flexible methanol synthesis

The synthesis at the level 2 starts with the optimal structure of the first level. Firstly, a group of critical vertices was determined by applying procedure described in 2.2.1.

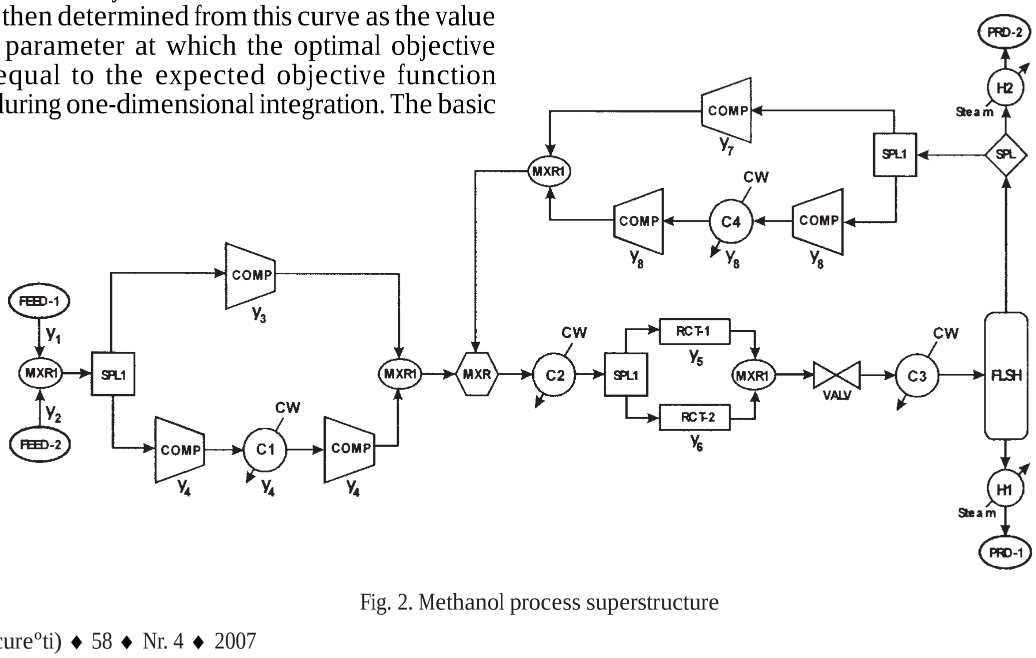

Fig. 2. Methanol process superstructure

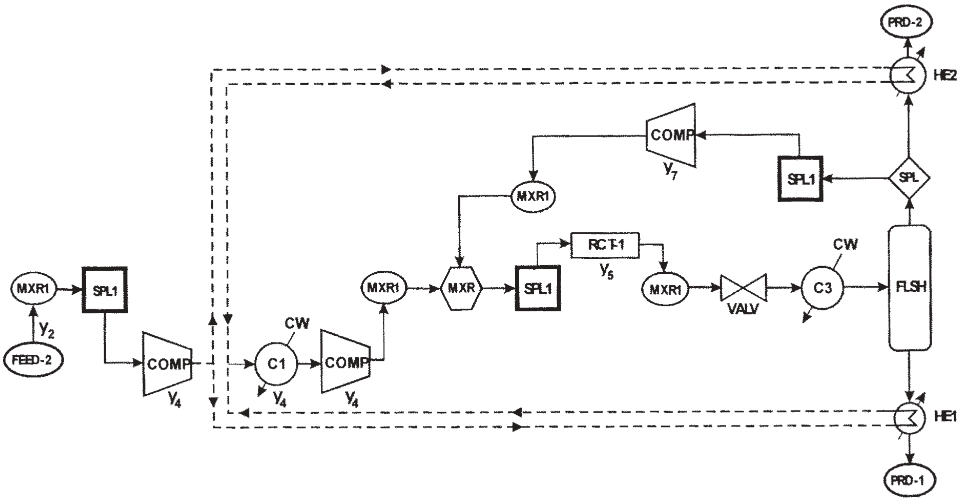

Fig. 3. Optimal non-flexible and flexible flow sheet topology

Table 1
SIZE OF THE MODELS USED IN THE FLEXIBLE METHANOL SYNTHESIS

	Level 1	Level 2	Level 3	Critical points determination	CBP determination
No. of points	1	5	5	1	9
Continuous variables	572	2656	2656	589	4740
Binary variables	46	46	46	/	/
(In)equality constraints	580	2892	2892	566	5204
CPU (s)	≈ 0.1 (per NLP) ≈ 0.1 (per MILP)	≈ 2.5 (per NLP) ≈ 0.85 (per MILP)	≈ 1.7 (per NLP) ≈ 0.6 (per MILP)	≈ 1.6 (per each structure)	≈ 187 (per each structure)

Table 2
VALUES OF DESIGN VARIABLES

	Level 1	Level 2	Level 3
Power COMP-2 (MW)	18.5	29.6	29.6
Power COMP-3 (MW)	15.5	28.0	28.0
Power COMP-4 (MW)	3.3	3.3	3.0
Volume RCT-1 (m^3)	72.8	77.4	77.9
Area HE1 (m^2)	558	529	529
Area HE2 (m^2)	208	403	401
Area Cooler C1 (m^2)	518	946	967
Area Cooler C3 (m^2)	2436	2397	2368

It was established that, in the case of the first structure, only four vertices constitute this group. The mathematical model was transformed into an indexed form and flexible synthesis was then performed simultaneously at the nominal point and at the critical points. Table 1 represents the sizes of all mathematical models used in the synthesis of flexible methanol process.

MIPSYN, an MINLP process synthesizer with a modified OA/ER algorithm [14] was used to perform MINLP iterations. No better topology than the first structure was found. However, the profit was significantly reduced when compared to the non-flexible solution, i.e. from 37.37 MUSD/yr to 33.04 MUSD/yr. This is mostly because of larger compressors on the feed stream, a larger reactor, and some exchangers.

Table 2 represents the values of design variables obtained at all three levels of applied approach. It is interesting to note, that not all the equipment in the flexible solution (Levels 2 and 3) are larger than the corresponding values of non-flexible design (Level 1); some units are also smaller. This is observed especially by heat exchanger network, where the total heat-transfer area of the flexible solution (4200 m^2) is higher than the total area of the non-flexible one (3720 m^2). However, some exchangers in flexible design are smaller than their counterparts in non-flexible network. Similar can be observed for total compression power.

Flexibility index was determined for the best flow sheet considering deviations of influencing uncertain parameters. The value obtained was 1.004, which indicates a flexible solution. Optimal design was further tested by applying Monte Carlo simulation over 4000 randomly selected points that assure the mean value within an error of ±0.23 MUSD/yr at 95 % confidence limit. The expected value obtained with Monte Carlo was 32.82 M$/yr which indicates that the nominal result (33.04) is within required confidence interval.

MINLP level 3 - Approximate stochastic flexible methanol synthesis

In this MINLP step, the normal distributions of uncertain parameters were defined with mean values equal to the nominal values and total deviation intervals equal to six-times standard deviations (6*s*). The central basic point was determined for the optimal structure obtained at the previous level and the synthesis started at this point, and at the critical points. The course of the OA/ER algorithm for levels 2 and 3 is presented in table 3.

Table 3
ITERATIONS OF OA/ER ALGORITHM (LEVELS 2 AND 3)

Iteration	NLP	MILP
LEVEL 2: starts from the optimal structure of level 1		
1	**33.04**	59.31
2	-3.97	-0.27
3	29.81	STOP
LEVEL 3: starts from the optimal structure of level 2		
4	**32.75**	59.28
5	STOP	

The approximated expected profit of the optimal structure is 32.72 MUSD/yr. The values of the design variables were close to those obtained using the nominal approach (table 2). Monte Carlo simulation yielded the expected profit of 32.81 MUSD/yr. This may indicate that, in the case of normal distributions of uncertain parameters and linear or moderately nonlinear objective functions, nominal point could give sufficiently accurate approximation of the expected value and exhaustive stochastic optimizations can thus be avoided.

Conclusions

A strategy is presented for the MINLP synthesis of flexible process flow sheets with many uncertain parameters. The procedure is evolutional and progresses from simple, less accurate steps to more demanding, but more accurate steps. The lower levels assure good initial structures for higher levels which then converge faster. Moreover, it is expected that in many cases optimal topology could be found at the lower levels, while at the upper level only fine adjustments of the design variables and the expected objective value are performed. A further motivation is thus to improve methodology in order to obtain flexible process flow sheets in just a few iterations.

A methanol case study has shown that simultaneous consideration of flexibility during the synthesis of optimal flexible flow sheet stimulates the establishment of several internal trade-offs, especially within some process subsystems, like heat exchanger networks and compression subsystems. The total sizes of such subsystems are certainly larger in flexible solutions than in non-flexible ones, however, individual process units could be either larger or smaller. This indicates that only simultaneous inclusion of operability issues in the process synthesis, in contrast to empirical oversizing of equipment, may establish the correct trade-offs and thus produce optimal solutions.

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References

1.GOEL, V., GROSSMANN, I. E., EL-BAKRY, A. S., MULKAY E. L., Comput. Chem. Eng., 30, nr. 6-7, 2006, p. 1076
2.RYU, J. H., DUA, V., PISTIKOPOULOS E. N., Comput. Chem. Eng., 28, 6-7, 2004, p. 1121
3.SAHINIDIS, N. V., Comput. Chem. Eng., 28, nr. 6-7, 2004, p. 971
4.DUA, V., PISTIKOPOULOS, E. N., Trans IChemE, Part A, 76, nr. A3, 1998, p. 408
5.ACEVEDO, J., PISTIKOPOULOS, E. N. Comput. Chem. Eng., 22, nr. 4-5, 1998, p. 647
6.AALTOLA, J. Appl. Therm. Eng., 22, nr. 8, 2002, p. 907
7.WEI, J., REALFF, M. J., Comput. Chem. Eng., 28, nr. 3, 2004, p. 333
8.BIEGLER, L. T., GROSSMANN, I. E., WESTERBERG, A. W. Systematic Methods of Chemical Process Design. Prentice Hall, New Jersey, 1999, p. 699
9.NOVAK PINTARIÈ, Z., KRAVANJA, Z. Proceedings on the ESCAPE-16. Elsevier, 21A, 2006, p. 503
10.NOVAK PINTARIÈ, Z., KRAVANJA, Z., Submitted to Comput. Chem. Eng., 2006
11.NOVAK PINTARIÈ, Z., KRAVANJA, Z., Comput. Chem. Eng., 28, nr. 6-7, 2004, p. 1105
12.KRAVANJA, Z., GROSSMANN, I. E., Comput. Chem. Eng., 14, nr. 12, 1990, p. 1363
13.YEE, T.F., GROSSMANN, I. E., Comput. Chem. Eng., 14, nr. 10, 1990, p. 1165
14.KRAVANJA, Z., GROSSMANN, I. E., Comput. Chem. Eng., 18, nr. 11/12, 1994, p. 1097

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