COMPARISON OF MATHEMATICAL PROGRAMMING AND PINCH-BASED TECHNIQUES FOR MASS EXCHANGE NETWORK SYNTHESIS

Z. SZITKAI, A. K. MSIZA[1], D. M. FRASER[1], E. RÉV, Z. LELKES and Z. FONYÓ

(Chemical Engineering Department, Budapest University of Technology and Economics, H-1521 Budapest, HUNGARY
[1]Chemical Engineering Department, University of Cape Town, Private Bag, Rondebosh, 7701, SOUTH AFRICA)

In this paper two design techniques, based on mathematical programming, for the synthesis of mass exchange networks (MENs) are compared. Problems not generally dealt with in literature, and associated with these techniques, are highlighted. A method is presented for generating several feasible initial solutions to avoid accepting poor local solutions as final designs. Methods of handling the discontinuity of the Kremser equation used for determination of the number of stages are also discussed. In addition, a method of generating MINLP (mixed integer non-linear programming) solutions that feature integer stage-numbers is also presented. It is shown that insight-based superstructures assuming vertical mass transfer may fail to include the optimal structure of the MEN. An MINLP based design technique is selected for the solution of several MEN synthesis example problems. Our MINLP solutions are compared to the advanced pinch solutions taken from literature. Both simple and advanced capital costing functions are used for the estimation of the total annual cost (TAC) of the MENs.

Keywords: mass exchange networks, mathematical programming, pinch technology

Introduction

Mass exchange networks (MENs) are systems of interconnected direct-contact mass-transfer units that use process lean streams or external mass separating agents to selectively remove certain components (often pollutants) from rich process streams.

The notion of mass exchange network synthesis (MENS) and a pinch-based solution methodology for the problem were presented first by El-Halwagi and Manousiouthakis [1,2,3]. Subsequently, Papalexandri et al. presented a mixed integer non-linear programming (MINLP) design technique for MENS [4]. Later, Hallale and Fraser extended the pinch design method by setting up capital cost targets ahead of any design [5,6,7]. Recently, Comeaux [8] presented an optimisation based design methodology where the notion of vertical mass transfer is used to develop a superstructure optimised by non-linear programming (NLP). Using optimisation-based techniques, capital and variable costs of the network can be optimised simultaneously even in cases of large and multi-component design problems.

Our objective is to compare the design methods above. MENS problems taken from literature will be solved. MENs got by the different methods will be evaluated against each other, and the design methods will be rated based on the properties of the resulting networks.

In the first part of the paper the NLP design method of Comeaux is compared to the MINLP method of Papalexandri. After dealing with practical problems of the mathematical programming methods, the last part of this paper presents MINLP solutions for thirteen example problems solved with pinch methodology by Hallale [7].

Comparison of two optimisation-based techniques

The optimisation (mathematical programming) -based approach of design or synthesis consists of three major steps. The first step is the development of a superstructure (a representation of alternatives of which the optimum solution is selected). The second step is the formulation of a mathematical program. The third one is the solution of the optimisation model. The values of the model variables in the optimal solution define the structure of the desired mass exchange network and its optimal operating parameters simultaneously. General reviews of the area are given by [9,10,11]. The major achievement of the mathematical programming approach is that it replaces the multi-step iterative pinch

Table 1 Total annual costs of the different solutions of Hallale's example problem 5.1

Solution method	Simple capital costing	Advanced capital costing
Advanced pinch method of Hallale and Fraser	*320,000 USD/yr. (target)	*228,000 USD/yr.
NLP method of Comeaux	*332,000 USD/yr.	249,150 USD/yr.
MINLP method of Papalexandri	306,108 USD/yr.	221,357 USD/yr.

*Original solutions of the referred authors are marked by an asterisk

Fig.1 Insight-based NLP solution for Hallale's example problem 5.1. Comeaux's method is used. The capital cost of the network is calculated by Hallale's advanced capital costing method

design method, and enables the design of multicomponent MENs as well. On the other hand, depending on the mathematical formulation, optimisation models cannot always be solved for global optimality [12].

In this section the MINLP method of Papalexandri [4] and the insight-based NLP solution of Comeaux [8] are compared. Both methods are mathematical programming based but differ in the way the superstructure is generated, and in the general classification of the mathematical programming model.

The superstructure of Papalexandri [4] contains all the imaginable matches (stream pairings) and bypasses, without considering whether a certain match is thermodynamically feasible or not. Then the superstructure is formulated as an MINLP problem, in which binary variables denote the existence of the matches. This method is simple in the sense that during the design process no preliminary knowledge about the network is needed. On the other hand, the resulting MINLP model may be superfluously large and hence difficult to solve. Commercially available MINLP solvers [13,14,15] may fail finding the global optimum of the problem.

Because of the mathematical difficulties, many efforts were made to combine thermodinamical insights and mathematical programming [8,16]. One of these attempts is the method of Comeaux [8]. Using stream data and the principle of vertical mass transfer an insight based superstructure is generated, which contains thermodinamically feasible matches only. Having cutted out many of the matches, the resulting superstructure and the corresponding mathematical program is much smaller, compared to that of Papalexandri. Comeaux's further simplification is omitting binary variables. He formulates MENS as pure NLP problems. Mass exchangers exchaging only a small amount of mass in the solution are regarded as non-existing ones. The pure NLP formulation (called the "cover and eliminate method" in another way) is one of the oldest optimisation based design concepts. Still, in may cases it can successfully be used, for example when solving wastewater treatment problems [17].

For the sake of a parctical comparison the MENS example problem 5.1 of Hallale [7] is solved using both of the methods outlined above. Since the selection of the objective function is a crutial point of the mathematical
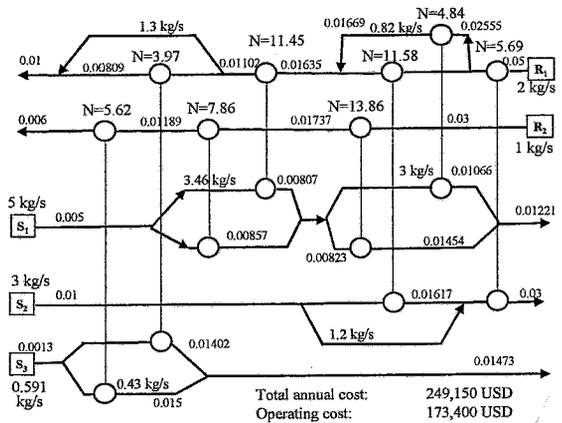
programming method, two different methods were used for calculating the total annual costs (TACs) of the networks. The two costing procedures differ in the way the capital costs are calculated. At first, a simple capital costing procedure is applied, where it is assumed that the capital cost of an exchanger (in USD/yr) can be calculated by multiplying its theoretical number of stages by 4552 (see Papalexandri et al., [4]). Secondly, the advanced, exchanger volume-based capital costing of Hallale and Fraser [5,6,7] is used. The latter one gives more realistic estimates for the capital and hence for the total annual costs.

Total annual costs (TAC) of the designed MENs can be found in Table 1. As a basis for further comparison, in Table 1 TACs of the supertargeted pinch solutions of Hallale [7] are shown as well.

Throughout this paper, optimisation problems are solved using the GAMS program package [13,14]. CONOPT-1 was used to solve the NLP models and DICOPT++ (running OSL and CONOPT-1) was used for the MINLP solutions.

The network obtained by the insight based NLP method using advanced capital costing is presented in Fig.1. Our MINLP designs based on Papalexandri's method can be seen in Figs.2 and 3 with simple and advanced capital costing structures, respectively.

Table 1 shows that the TAC of the MINLP designs and the advanced pinch solutions are approximately the same. This is seen more clearly when the structures of these solutions are compared. The costs also become closer when rounding up the stage numbers of the MINLP solutions (to $322 000 and $226 000 respectively). Table 1 also shows that Comeaux's method gives more expensive solutions. Hallale's design (see Fig.5.4 of Hallale [7]) cannot be reproduced by using the insight-based NLP method of Comeaux. This is because the insight based superstructure for the example (see Fig.4.2 of Comeaux [8]) does not enclose the structure of Hallale's solution. In order to be able to find Hallale's solution, additional possible matches should be added to the superstructure generated using the principle of vertical mass transfer. Although the
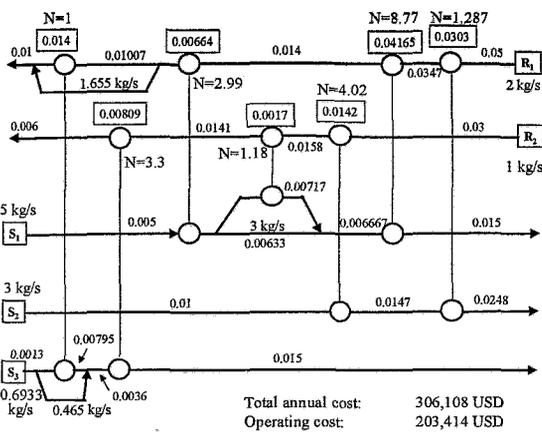
Fig.2 MINLP solution of Hallale's example problem 5.1. The solution is obtained by Papalexandri's method, using a simple capital cost correlation


Fig.3 MINLP solution of Hallale's example problem 5.1. The capital cost of the network is calculated by the advanced capital costing method of Hallale and Fraser. Papalexandri's MINLP method is used

NLP-based design method is computationally simpler than the MINLP method, it lacks the advantages of the latter. Logical conditions for fixed costs, integer stage numbers, etc. cannot be handled in an NLP model.

Because of the reasons outlined above, authors of this article decided to use Papalexandri's MINLP method for the synthesis of MENs.

Three practical problems of using optimisation-based design techniques for mass exchange network synthesis

Generation of feasible initial solutions

Global optimality for a given MINLP solution by GAMS/DICOPT (the outer approximation algorithm) is guaranteed when both the objective function and the feasibility region of the MINLP model are convex. The feasibility region of the MINLP problem is convex when all the equalities consist of linear functions and the inequalities consist of convex functions [12,11]. In most cases however, these conditions cannot be satisfied, or they can be satisfied only by oversimplifying the physico-chemical model of the design problem. For example, the MINLP or NLP models for MEN synthesis problems contain non-linear equality constraints (mass balances, phase equilibria), non-convex equations (e.g. the Kremser equation), as well as non-convex terms in the objective function (e.g. typical cost functions like $cost = const \cdot (size)^{0.6}$, etc.). Non-linear equalities, non-convex inequalities and a non-convex objective function can be handled using convexifying techniques [18] or piecewise linearisation [9]. Using these techniques, many additional binary and continuous variables have to be included into the MINLP model, and this way the problem size increases steeply. In general we can state that non-convexity is an inherent property of rigorous MINLP models for process design. Though the MINLP solver GAMS/DICOPT++ can handle nonconvexities to some
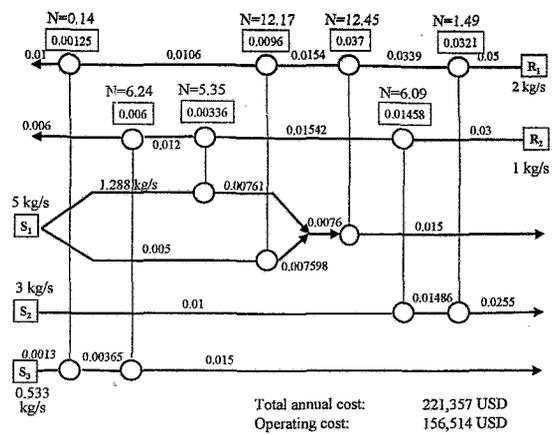
extent, global optimality is not guaranteed for the solution [14].

In consequence of the above, solutions obtained from different initial values have to be compared to prevent the selection of poor local solutions as final designs. Once a feasible solution is found starting from a particular initial solution, several other feasible initial values can be generated, by changing the objective function of the original problem, or by adding artificial constraints. A solution featuring minimum MSA flowrates can, for example, be used as the initial solution for the original problem. Simple pinch solutions can also serve as initial solutions. If the problem is not severely non-convex, the initial feasible solution can be obtained from any nonzero set of initial values for the variables in the model.

Having several initial values on hand, the solution of the original NLP or MINLP problem is started from all of them, now using the original objective function. The best solution with the lowest total annual cost is selected as the final solution. Though this procedure involves the elements of a "trial and error" method, it performs well in the computational practice. Since there is no algorithm, which could deliver the globally optimal solution for a non-convex MINLP problem, this is a reasonable way of calculation when using commercially available MINLP solvers.

The discontinuity problem of the Kremser equation

Assuming linear phase equilibrium relations, capital investment calculations of the mass exchangers (absorbers, extractors) are most commonly based on the Kremser equation [19], which gives the required number of equilibrium stages for a given separation task. Depending on the value of the removal factor A, the Kremser equation for a given component i has two different forms.

If $A \neq 1$ then

$$N_{A \neq 1} = \frac{\ln\left(\left(1 - \frac{1}{A}\right)\left(\frac{y_i^{in} - m_j x_j^{in} - b_j}{y_i^{out} - m_j x_j^{in} - b_j}\right) + \frac{1}{A}\right)}{\ln(A)} \quad (1)$$

where

$$A = \frac{L_j}{m_j G_i} \quad (2)$$

If A=1 then

$$N_{A=1} = \frac{y_i^{in} - y_i^{out}}{y_i^{out} - m_j x_j^{in} - b_j} \quad (3)$$

In the formulas above y_i^{in}, y_i^{out} denote the inlet and outlet concentrations of the ith rich stream, x_j^{in}, x_j^{out} denote the inlet and outlet concentrations of the jth lean stream, and L_j, G_i denote the lean and rich flow rates. The equilibrium relation between the ith rich stream and the jth lean stream is given in the form of $y_i^{out} = m_j x_j^{in} + b_j$.

The removal factors of the units are design variables when solving the MINLP synthesis problem. Because of this, the A values have to be able to vary freely between their possible bounds. The A value can be either greater, less or equal to the value 1. The theoretical number of stages as a function of the removal factor A have a removable discontinuity at $A=1$. In case of calculating the number of theoretical stages at $A=1$, $Eq.(3)$ has to be used. Using only the first form of the Kremser equation ($Eq.(1)$) usually leads to a division by zero error or provides a solution that has no physical meaning. To be more precise, the solver sets as many A values to 1 as possible because at $A=1$ the number of theoretical stages can be zero, independently from the concentrations (See $Eq.(1)$). Restricting the values of A under or over 1 excludes the real optimal solution from the search space. No reasonable MENs can be expected from the MINLP method without solving this numerical difficulty.

How the discontinuity of the Kremser equation can be handled when using commercially available MINLP solvers, is extensively discussed in our previous papers [20,21].

When the MENS problem is formulated as a pure NLP problem, binary variables cannot be used for handling the discontinuity of the Kremser equation. For the pure NLP case here we present a method [2"], which is an approximation, still it can be used effectively. $Eq.(4)$ for all of the mass exchangers can be introduced:

$$A - 1 = (A - 1 + \varepsilon) \cdot w \quad (4)$$

where A is the absorption factor of the mass exchange unit, and ε is a small positive number (eg. 0.001). The variable w takes the value of zero when $A=1$, and is close to one when $A \neq 1$. Then $Eq.(5)$ is used in the model for choosing between the two stage numbers (See also $Eqs.(1)$ and (3)).

$$N = (1 - w) \cdot (N_{A=1}) + w \cdot (N_{A \neq 1}) \quad (5)$$

Using this method, the total cost of the network must be recalculated after optimisation. Experience has shown that differences between the real and the approximated stage numbers are usually small. It has to be noted that $Eq.(5)$ is a bilinear equality constraint, hence it renders the NLP problem nonconvex.

An obvious solution for the Kremser discontinuity problem could be the introduction of $Eq.(6)$, which prevents A from taking the value of unity:

$$(A - 1)^2 \geq \varepsilon \quad (6)$$

Using this constraint only $Eq.(1)$ should be used, when calculating the theoretical number of stages of the units. According to our computational experience however, this method leads to severe numerical problems during the solution of the NLP, and therefore cannot be applied.

Integer stage numbers

The designer may often want to get solutions whereby the number of stages (N) are reported as integers. Buliding up the integer-values according to $Eq.(7)$ requires the introduction of additional binary variables (z_i) in the MINLP model.

$$N = 2^0 \cdot z_1 + 2^1 \cdot z_2 + 2^2 \cdot z_3 + \ldots \quad (7)$$

When the expected number of stages for the exchangers in the superstrucure is large, the additional binary variables may extend the MINLP problem size over the solvability limit. The same method however, can be used in a two-level optimisation approach. After obtaining the solution with non-integer stage numbers, a second optimisation can be carried out, where the structure of the network is now fixed to that of the first solution (with non integer stage numbers). In the second level only the operating parameters of the network and the stage numbers around the first solution are optimised. For example, if an exchanger in the first level optimum has 16.3 stages then in the second level $Eq.(8)$ is introduced, allowing the number of stages of that particular unit to change between 15 and 18:

$$N = 15 + 2^0 \cdot z_1 + 2^1 \cdot z_2 \quad (8)$$

In this way fewer binary variables are needed. However, the two-level method fails to find the optimum when the rounding affects the optimal structure of the network.

Solved MENS example problems

In section two it has been shown that in spite of its difficulties, the MINLP method of Papalexandri [4] is still better applicable than the NLP optimisation of Comeaux's insight based superstructure [8]. In section three some important practical problems of the optimisation based MENS have been identified, and

Table 2 Comparison of our MINLP solutions and the advanced pinch solutions of Hallale [7]. CAP indicates that the network was optimised only for its capital cost at fixed lean stream flow rates (operating costs). Theoretical number of stages, hence capital costs are rounded up where staged vessels are used

Example	Objective function	Pinch solution of Nick Hallale Target / Design	Our MINLP Solution	$100*(C_{MINLP}-C_{Pinch})/C_{MINLP}$
3.1	CAP	830 000 / 860 000	1 044 285	+17.6 %
3.2	CAP	448 000 / 455 000	453 302	-0.4 %
3.3	CAP	819 000 / 751 000	637280	-17.8 %
3.4	CAP	591 760 / 637 000	637 000	0.0 %
4.1	CAP	296 000 / 298 000	255 068	-16.8 %
5.1	TAC	226 000 / 228 000	226 000	-0.9 %
5.2	TAC	226 000 / 228 000	226 000	-0.9 %
5.3	TAC	226 000 / 228 000	226 000	-0.9 %
5.4	TAC	49 000 / 49 000	50 279	+2.5 %
5.5	TAC	524 000 / 526 000	527 000	+0.2 %
6.1	TAC	692 000 / 706 000	720 000	+1.9 %
6.2	TAC	28 000 / 28 000	32 000	+12.5 %
6.3	CAP	591 000 / 539 000	536 000	-0.6 %

TAC – total annual cost in USD/yr, **CAP** – annualised capital cost in USD, **C** – cost

annual capital cost of this MINLP design: 615 287 USD
when rounding up the stage numbers: 637 280 USD

Fig.4 MINLP solution of Hallale's example problem 3.3. Papalexandri's MINLP method and simple capital costing are used

this MINLP design: 255,068 USD
N. Hallale's solution: 298,000 USD

Fig.5 MINLP solution of Hallale's example problem 4.1. Papalexandri's MINLP method and advanced capital costing are used. Mass exchangers are packed columns here

solutions for these problems were presented, too. Now we are in position to solve MENS problems that had already been solved in literature using Pinch technology. In this section MINLP solutions for most of the MENS example problems of the thesis of Hallale [7] are presented.

Our results are summarised in *Table 2*. The first column shows the reference number of the examples in Hallale's thesis. The second column of the table indicates the type of the objective function. Some of the pinch solutions are optimised only for the capital cost of the exchangers at fixed mass separating agent (MSA) flow rates. The objective functions of these examples are indicated by CAP (capital cost), because the fixed MSA flow rates define the variable costs of the networks. The costs of the pinch targets and final designs are shown in column three, while the costs of our MINLP solutions can be found in the fourth one. Theoretical number of stages, hence capital costs are rounded up where staged vessels are used. Column five shows how many percents the cost of our MINLP solution is cheaper or more expensive than that of the pinch solution.

Optimising for TAC, in most cases equally good solutions are obtained. Total annual costs of the MINLP and pinch solutions do not really differ from each other. In case of examples 5.1, 5.2, 5.3 the MINLP solutions, in case of examples 5.4, 5.5, 6.1 the pinch solutions prove to be a bit cheaper. The only exception is example 6.2 where the advanced pinch solution is 12.5 % cheaper than the MINLP solution. The difference in this particular case can be accounted for the fact that the concentration range of this example extends over six orders of magnitude. The concentration variables cannot be scaled. Having great differences in the values of the model variables causes numerical difficulties for the MINLP solution algorithm.
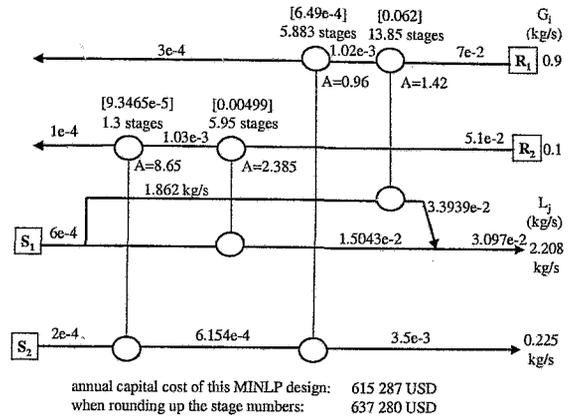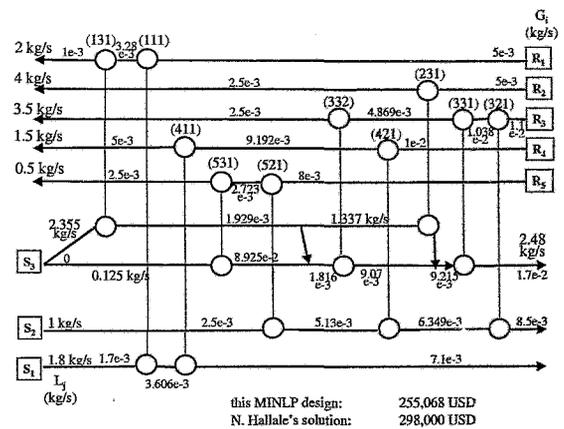
Optimising just for the annualised capital cost (CAP), usually better or equally good MINLP solutions are obtained. Out of the six examples, the MINLP and pinch solutions are almost the same in the cases of examples 3.2, 3.4, 6.3. MINLP solutions of examples 3.3, 4.1 are significantly better (17.8 % and 16.8 % cheaper) than the corresponding pinch solutions, while in the case of example 3.1 the advanced pinch solution proved to be 17.6 % cheaper.

Why the pinch solutions are not always reached with MINLP can be accounted for the inherent nonconvexity of the problem. Though in many cases solutions can be improved by starting the calculations from different initial values, it is not guaranteed at all that better (cheaper) solutions can always be found. A good illustration for this is the MINLP solution of example 3.1.

As our solutions show, the advanced pinch targeting method of Hallale and Fraser delivers a good estimate for the capital and total annual costs. Nevertheless, it has to be noted that the pinch targets are not rigorous,

since in the case of examples 3.3 and 4.1, using MINLP even the targeted costs could be surpassed in design. The MINLP solutions of examples 3.3 and 4.1 are shown in *Figs.4* and *5* respectively.

Based on the experience, obtained during the solution of the thirteen single component example problems we conclude that the time consuming, heuristic pinch design method can be replaced by the MINLP design concept. Nevertheless, knowing the drawbacks of the methods, it is advisable to consider both methodologies when solving a problem.

Conclusions

The MINLP design method of Papalexandri seems to be superior to the NLP optimisation of insight-based superstructures. It is shown that insight based supersructures constructed using the vertical mass transfer principle may not enclose the optimal structure of the mass exchange network. The practical problems which were identified in optimisation-based MENS, namely generation of feasible initial solutions, discontinuity of the Kremser equation and integer stage numbers can effectively be handled, as described in Section 3. Based on the experience, obtained during the solution of thirteen single component example problems we conclude that the time consuming, heuristic pinch design method can be replaced by the MINLP design technique.

REFERENCES

1. EL-HALWAGI M. M. and MANOUTHIOUSAKIS V.: AIChEJ, *1989*, 8, 1233-1244
2. EL-HALWAGI M. M.: "Pollution Prevention Through Process Integration", Academic Press 525 B Street, Suite 1900, San Diego, California 92101-4495, USA, (www.apnet.com) 1997
3. EL-HALWAGI M. M. and MANOUSIOUTHAKIS V.: Chem.Eng.Sci., *1990*, 45(9), 2813-2831
4. PAPALEXANDRI K. P., PISTIKOPOULOS E. N. and FLOUDAS C. A.: Trans IChemE, *1994*, 72, Part A, 279-293
5. HALLALE N. and FRASER D. M.: Chem. Eng. Science, *1998*, 53(2), 293-313
6. HALLALE N. and FRASER D. M.: Trans I Chem E, *2000*,78, Part A, p.202-216
7. HALLALE N.: Capital Cost Targets for the Optimum Synthesis of Mass Exchange Networks, PhD thesis, University of Cape Town, Dept. of Chemical Engineering, 1998
8. COMEAUX R. G.: Synthesis of MENs With Minimum Total Cost, MSc Thesis, Dept.of Process Integration, UMIST, Manchester, England, 2000
9. BIEGLER L. T., GROSSMANN I. E. and WESTERBERG A. W.: "Systematic Methods of Chemical Process Design", Prentice Hall International Series in the Physical and Chemical Engineering Sciences, 1997
10. GROSSMANN I. E.: MINLP optimization strategies and algorithms for process synthesis. Proc. of FOCAPD'89, Snowmass, Colorado, 1989
11. DURAN M. A. and GROSSMANN I. E.: AIChE Journal, *1986*, 32, 592
12. DURAN M. A. and GROSSMANN I. E.: Math. Program *1986*, 36, 307
13. BROOK, KENDRICK, MAEREUS: GAMS A User's Guide, Release 2.25, Scientific Press, 1992
14. GAMS Development Corporation, 1217 Potomac Street Washington, DC 20007, USA, "GAMS-The Solver Manuals, GAMS/DICOPT", (www.gams.com)
15. SCHWEIGER C. A. and FLOUDAS C. A.: "MINOPT: A Modeling Language and Algorithmic Framework for Linear, Mixed-Integer, Nonlinear, Dynamic, and Mixed Integer Nonlinear Optimization", Princeton University, (http://titan.princeton.edu/MINOPT) 1998
16. HOSTRUP M., GANI R., KRAVANJA Z., SORSAK A. and GROSSMANN I. E.: Computers and Chemical Engineering, *2001*, 25, 73-83
17. BENKŐ N., RÉV E.,SZITKAI Z. and FONYÓ Z.: Computers Chem. Engng., *1999*, 23, Supp. pp. S157-S160
18. PÖRN R., HARJUNKOSKI I. and WESTERLUND T.: Computers Chem. Engng., *1999*, 23, pp.439-448
19. KREMSER A.: Natl.Petrol.News, *1930*, 22(21), pp. 42
20. SZITKAI Z., LELKES Z., RÉV E. and FONYÓ Z.: Solution of MEN synthesis problems using MINLP: Formulations of the Kremser equation, ESCAPE-11, Elsevier, 2001
21. SZITKAI Z., LELKES Z., RÉV E. and FONYÓ Z.: „Handling of removable discontinuities in MINLP models for process synthesis problems, formulations of the Kremser equation", accepted for publication in Computers and Chemical Engineering., 2002, in press
22. SZITKAI Z., MSIZA A. K., FRASER D. M., RÉV E., LELKES Z. and FONYÓ Z.: "Comparison of different mathematical programming techniques for mass exchange network synthesis", Escape-12 proceedings, in press