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Abstract

This paper deals with the global optimization of networks consisting of splitters, mixers and linear process units and which involve multicomponent streams. Examples include pooling and blending systems and sharp separation networks. A reformulation-linearization technique is first applied to concentration and flow based models in order to obtain a relaxed LP formulation that provides a valid lower bound to the global optimum. This formulation is then used within a spatial branch and bound search. The application of this method is considered in detail for sharp separation systems with single feed and mixed products. Numerical results are presented on twdve test problems to show that only few nodes are commonly required in the branch and bound search.

Introduction

A common source of noneonvexities in the synthesis and design of processes, as well as in flowsheet optimization, are the material flow equations for multiccmponent streams. These nonconvex equations involve bilinear terms and they arise in the mass balance equations when the compositions are unknown. There are different equivalent formulations for this type of networks. One alternative is to formulate the mass balance equations in terms of component concentrations. In this form bilinear terms are present in the equation* for the mixer units and the different process units (e.g. sharp separators). A second alternative is to express the mass balances in terms of flows of individual components. This option has the advantage that it involves a ^naUmr number of nonlinear equations. However, the modelling of the splitter units involves bilinear terms that arise due to the condition that the proportions of flows between components have to be the same for the different streams.

The difficulty with the nonconvexities noted above is that they may give rise to optimization problems Involving several local optima and numerical singularities that may produce failure in the NLP algorithms. Recently there have been important efforts in the area of global optimization. Examples of algorithms are the ones proposed by McCormick (1976). Floudas and Viswewaran (1990) and Sherali and Alameddine (1992) which can be used to solve bilinear programming problems like the ones that arise in networks with multicomponent streams. For a recent review in the area of bilinear programming see Al-Khayyal (1992).

As for previous work in the design and synthesis of multicomponent process networks Mahalec and Motard (1977) and Nath (1977) developed evolutionary techniques that are based on heuristics to generate a network configuration. Floudas (1987) addressed the synthesis of separation networks with mixed products in which only sharp separators are considered. A superstructure of the process network was proposed and modelled in terms of concentrations. The resulting model is nonconvex and solved with a standard NLP algorithm with no guarantee of global optimality. Floudas and Aggarwal (1990) solved small pooling and blending problems and sharp separation networks problems using a strategy based on Benders decomposition. In this approach only convex subproblems are solved but there is no guarantee of obtaining the global optimum. Kocis and Grossmann (1989) modelled process networks with multicomponent streams in terms of the individual component flows. They included a set of bounding constraints with binaxy variables to approximate the nonconvexities that are present in splitters with multiple outputs. Wehe and Westerberg (1987) studied the problem of sharp separation networks with mixed products. They proposed a search procedure that involves the

enumeration of the different separation sequences. The nonconvex equations are dropped and constraints that are valid for each particular sequence with a set of bounds over the key components are included to obtain tighter UP relaxations for each configuration. However, the number of sequences to be examined grows rapidly and there is no guarantee of global optimality.

In some particular cases the nonconvexities in the mass balances can be avoided through the introduction of binary variables. One of these cases is when single choice splitters are present in the flowsheet (Kocis and Grossmann, 1989). Here, it is possible to have a mixed integer linear formulation for the mass balance equations in terms of the individual component flows. Another restricted case for which some nonconvexities can be reformulated is when mixing within the network is only allowed for streams of the same concentration. In this form, larger network superestructures must be proposed and the concentrations of the streams are known beforehand. Integer variables are introduced to model the existence of the different streams (e.g. the mixed integer linear formulation for sharp separation networks by Floudas and Anastasiadis, 1988).

The objective of this paper is to present an efficient global optimization method that exploits the particular structure that is present in process networks with multicomponent streams (e.g. pooling and blending systems, sharp separation networks). First a relation is established between formulations based on concentrations and individual flows. This is done following the Reformulation-Linearization technique proposed by Sherali and Alameddine (1992). As will be shown, a linear relaxation Is obtained that is in the space of the concentrations and individual flows which can be used in a branch and bound search is to find the global optimum. Application to the optimal design of sharp separation systems with single feed and mixed products is considered In detail. Different preprocessing techniques that allow tightening of the relaxation problem are presented. The performance of the algorithm is reported on a total of twelve problems.

Modeling with concentrations and individual flows

Consider a process network that consists of splitters, mixers and process units that are interconnected with multicomponent streams (see Fig. 1). The process iinits that are considered in this paper are units in which the output flows of the components can be expressed as a linear relation of the inlet flows (e.g. sharp separators, reactor with^known conversion). It is possible to formulate the mathematical model of the process network in terms of the concentrations of the streams. Xj^k. Another possibility is to model the network

using flows of individual components. The former has the advantages that it provides a convenient framework for the evaluation of therxnodynamic properties, and in many cases bounds can be expressed in a more natural form. A major disadvantage is that many nonconvex terms (bilinear) are involved in the mass balances for the components. The individual component flows formulation is often chosen since it gives rise to a larger number of linear equations and the only nonconvexities are involved in the modelling of the splitters. In these untis it is necessary to enforce that the components maintain the same concentration in each of the streams leaving the splitter. These constraints can be expressed as relations between the different components (Wehe and Westerberg, 1987). One deficiency of this representation is that since many flows can take values of zero, singularities may arise with which conventional nonlinear programming methods may have difficulties to converge. Another alternative is to introduce additional variables that represent split fractions (Kocis and Grossmann. 1989). This involves a larger number of constraints but tends to yield a formulation that numerically is better behaved.

Following axe the equations that apply to the mixers, splitters and units using the two alternative representations:

Mixer

A mixer k consists of a set of Inlet streams, M_k, and an outlet stream k (see Fig.2).

a) Concentrations

The total mass balance for a mixer k is given by:

$$F^k = \sum_{i \in M_k} F^i \qquad (1)$$

where F^i is the total flow in stream i. The mass balance for each component j is given by the nonlinear equations.

$$F^k x_j^k = \sum_{i \in M_k} F^i x_j^i \qquad \text{for all } j \qquad (2)$$

where x_j^i is the concentration of component j in the stream i

b) Individual flows

Here it is only necessary to write a mass balance for each component j, given by the linear equations:

$$f_j^k = \sum_{i \in M_k} f_j^i \qquad \text{for all } j \qquad (3)$$

where f_j^i is the flow of component j In stream i.

Splitter

A splitter k has an inlet stream k and a set of outlet streams S_k (see Fig. 3).

a) Concentrations

The equations for a splitter in terms of the concentrations are given by the following linear equations

$$\sum_{i \in S_k} P^i = P^k \qquad (4)$$

$$x_j^i = x_j^k \qquad \text{for all } i \in S_k \text{ and } j \qquad (5)$$

$$\sum_j x_j^i = 1 \qquad (6)$$

b) Individual flows

The mass balance for each component j is given by

$$\sum_{i \in S_k} f_j^i = f_j^k \qquad \text{for all } j \qquad (7)$$

Here, it is also necessary to enforce the condition that the streams leaving the splitter have the same proportions in flow for each component. These relations between components, which are nonlinear, can be expressed in terms of the inlet stream k and a given component j'

$$f_{j'}^k f_j^i = f_j^k f_{j'}^i \qquad \text{for all } i \in I_k \text{ and } j \neq j' \qquad (8)$$

A different approach consists of introducing as additional variables the split ratios ξ_t^f that represent the part of the inlet flow that goes to the outlet stream i. The nonlinear equations are given by

$$f_j^i = \xi^i f_j^k \qquad \text{for all } i \in I_k \text{ and } j \qquad (9)$$

with $0 \le \xi \le 1$.

Process units

In this paper it will be assumed that the outlet streams, $i \in O_k$, in the process units can be expressed as linear relations of the inlet streams, $i \in I_k$ (see Fig. 4). This is for instance the case of sharp split separators, separations in which the recovery level is known, or reactors that have a fixed conversion.

a) Concentrations

The overall mass balance for process unit k is given by,

$$\sum_{i \in I_k} P^i = \sum_{i \in O_k} P^i \qquad (10)$$

The mass balance for each component j is given by the nonlinear equations;

$$\sum_{i' \in I_k} \beta_{jr}^{ik} F^{i'} x_j^{i'} = P^i x_j^i \qquad \text{for all } i \in O_k \text{ and } j \qquad (11)$$

where P_{jr}^{ik} is a constant for process unit k that gives the distribution of component j in the stream $i \in O_k$ coming from streams $i' \in I_k$. For a separator unit it is required that $\sum_{i \in O_k} \beta^{ik} = 1$ and $\sum_{i' \in I_k} P_{jr}^{ik} = 1$. A sharp split separator is one for which $|I_k| = 1$ and $|O_k| = 2$ (top and bottom streams) and for all the components the constant p_{jr}^{ik} are either 0 or 1.

b) Individual flows

Only the mas balance for each component is necessary and it is given by:

$$f_j^i = \sum_{i' \in I_k} \beta_{ji'}^{k} f_j^{i'} \qquad \text{for all } i \in O_k \text{ and } j \tag{12}$$

A model in terms of individual flows MF consists of the linear equations (3), (7) and (12) plus the nonlinear equations (8) or (9). The model in terms of the concentrations, MX, includes the linear equations (1), (4), (5), (6) and (10) plus the nonlinear equations (2) and (11).

Reformulation and Linearization

In order to avoid the direct use of the nonconvex models MX and MF, there is a relation that can be established between them using the reformulation and linearization technique for bilinear programming models proposed by Sherali and Alameddine (1992). This technique can be applied to the model MX. First, consider the bounds over the variables present in the bilinear terms (total flow, F^i and concentrations x_j^i)

$$F^{iL} \le F^i \le F^{iU} \tag{13}$$

$$x_j^{iL} \le x_j^i \le x_j^{iU} \tag{14}$$

Using the bounds in (13), (14) the following constraints can be generated for the bilinear terms in (2) and (11),

$$F^i x_j^i \ge F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \tag{15}$$

$$F^i x_j^i \ge F^{iU} x_j^i + x_j^{iU} F^i - F^{iU} x_j^{iU} \tag{16}$$

$$F^i x_j^i \le F^{iU} x_j^i + x_j^{iL} F^i - F^{iU} x_j^{iL} \tag{17}$$

$$F^i x_j^i \le F^{iL} x_j^i + x_j^{iU} F^i - F^{iL} x_j^{iU} \tag{18}$$

In fact, McConnick (1976) has shown that the constraints in (15)-(18) correspond to the convex and concave envelopes of the bilinear terms over the given bounds. The formulation is linearized by the definition of the following variables:

$$f_j^i = F^i x_j^i \tag{19}$$

Tile resulting model which involves equations UK (3). (4). (5), (6). (10). (12) and the constraints in (15M18) is a linear relaxation of the original nonconvex concentration model, MX, in which the nonlinear equations (2) and (11) have been replaced by the linear equations (3) and (12) from the individual flow model. MF. It is possible to generate additional linear constraints that are redundant to the original nonlinear model, MX, but that can be nonredundant in the linear relaxation of the model (Sherali and Alameddine, 1992: Sherali et

al*. 1992). In particular, consider equation (7) that is the linear component mass balance for the splitters in model MF. "This linear equation is not present in the linear relaxation of the concentration model. MX. Take equation (4) and multiply by the valid bound constraint x,^{k} £ 0 to get

$$\sum_{i \in S_k} F^i x_j^k = F^k x_j^k \qquad (20)$$

Using equation (5) yields.

$$\sum_{i \in S_k} F^i x_j^i = F^k x_j^k \qquad (21)$$

that can be linearized to.

$$\sum_{i \in S_k} f_j^i = f_j^k. \qquad (22)$$

yielding equation (7). Hence, the linear equation for the splitter is valid and it is included. The nonlinear equations (8) or (9) can also be generated in a similar fashion but their linearizations are in general redundant (see Appendix A). They are only useful when the formulation 6f the problem provides non-trivial bounds over certain components in the outlet streams of a splitter, or when there exist some restrictions over the split ratios for the outlet streams.

Also, the constraints that relate the total flow and the individual flows of a stream can be generated for the splitters. Taking equation (6) and multiply by F^1 yields.

$$F^i \sum_j x_j^k = F^i \qquad (23)$$

Using the constraints $x_j^i = x_j^k$ in equation (23) and linearizing with $f_j^i = F^i x_j^i$ yields,

$$F^i = \sum_j f_j^i \qquad (24)$$

Based on the above it is possible to obtain a reformulated model MR that involves concentrations, total flows and component flows, and which bounds the solution of the original problem. The following equations are given for model MR:

a) Objective function. Φ which is expressed in terms of individual or total flows.

b) Mixer equations, which are expressed in terms of the total and individual component flows.

$$F^k = \sum_{i \in M_k} F^i \qquad (1)$$

$$f_j^k = \sum_{i \in M_k} f_j^i \qquad \text{for all } j \qquad (3)$$

c) Splitter equations, that are expressed in terms of the individual component flows and the f^oncditi^ tf^nff of the streams

y p-pk (4)
t€S_k

Xj' = Xj^k for alii *e* 3* and J (5)

XxJ<=i (6)

$_{i \in S_k}$£ If «f f for ail j (7)

d) Process units equations, that are given in terms of the total and individual component flows

$\sum_{i \in I_k} F^i = \sum_{i \in O_k} F^i$ (10)

fj«= $_{r \in I_k}$T p/'f/ foraUie $O_k$andj (12)

e) Relation between the total flow and the individual component flows

F^i = £ f/ for all streams (24)
j

f) Linear estimators, relate the individual component flows with the total flow and concentrations.

ri £ piLjqi + x* F* pi . piLjriL (15)

fji ^F^{1tt}^^{4}^^!^{u}F^l-F^{lu}^^{lu} for alii e S_k (16)

f/ ≤F^{ltt}^^{l}4-^^{1L}F^i-F$^{!u}$^^{IL} i s k € splitters flT)

fj^1 ^F^XJ' + X^F-F'-XJ' " foralljeJ (18)

f) Bounds on flows and concentrations

$F^{iL} \le F^i \le F^{iu}$ (13)

$x_j^{iL} \le x_j^i \le x_j^{iu}$ (14)

In previous approaches (Wehe and Westerberg, 1987; Kocis and Grossmann, 1989) looser approximations of the nonlinear terms were used. In both cases, the nonconvex problem (MF) was relaxed to a linear model by dropping the nonlinear equations (8) or (9). Equations that approximate the difference relation between the components were considered (Kocis and Grossmann, 1989). They were based on the difference that exists at the inlet of the splitter between the flowrate of the components and required the introduction of binaiy variables.

Outline of global optimization method

Model MR can be applied to predict lower bounds to the global optimum in the optimization of pooling and blending problems and in the synthesis of separation systems. The reason is that model MR provides a valid relaxation of the original feasible region since the nonlinear equations (2) and (11) in model MX are not considered, and the valid linear equations (3), (7), (12) and (15)-(18) are included. The proposed global optimization algorithm relies on the solution of the relaxed problem MR within a spatial branch and bound enumeration. The outline of the algorithm is as follows (for a more detailed description of step 4 see Quesada and Grossmann (1993))

0. Preprocessing (optional)

Determine bounds on the variables involved in the nonconvex terms, that is total flows, F^1, and concentrations, $x{$. Apply any additional preprocessing specific to the structure of the problem in order to further bound or flx variables.

1. *Lower Bound*

Solve model MR over a given subregion (initial subregion is the complete feasible region) minimizing a convex objective function $. If Φ is linear the model Is an LP.

2. Upper Bound

Any feasible solution to the nonlinear model provides an upper bound. Heuristic techniques can be employed to obtain good feasible solutions or the original problem, MF. can be solved using the solution of model MR as a good initial point. If the solution of problem MR is feasible it provides an upper bound.

3. *Convergence*

If the lower bound of a subregion is sufficiently close or above the upper bound discard that subregion. If no subregions are left the global solution corresponds to the best upper bound.

4. Branch and Bound

Partition the remaining subregions into a set of disjoint subproblems. Repeat steps 1-3 for each of the new subregions.

Remarks

The preprocessing step plays an important role in the above algorithm. It is during this step that initial bounds for the variables involved in nonconvex terms are obtained. The quality of

these bounds affects the tigthness of the lower bound since they are part of the estimator equations (15)418). Additionally, these bounds affect the performance of the algorithm because they define the search space over which the branch and bound procedure may have to be conducted.

In some cases, as described later in this papier, it is possible to exploit the particular structure of the process network and generate bounds for the variables without having to solve any subproblems. Furthermore, during this preprocessing step additional constraints can be generated for predicting a tighter lower bound of the global optimum can be obtained.

Some of the linear mass balances and the estimator equations are redundant in the nolinear formulations, MF and MX. These equations become nonredundant in the reformulated model, MR. and for that reason it is important to write a complete formulation of the network. However, this model can present some redundancies that can be easily identified and eliminated to reduce the size of the model. This is the case for the concentration variables used in the splitters. Model MR uses different sets of concentrations variables for the inlet and outlet variables of a split unit. In practice, it is only necessary to define the concentration of the component in the splitter and use the same variables for all the splitter streams. Also, some redundancies can occur with the total flow variables. These ones are necessary for the streams in the splitters but they might be redundant and eliminated in the other untis if they do not appear in other part of the model or in the objective function.

If the solution of model MR is feasible for the original nonconvex problem then it corresponds to the global optimal solution. When the solution to the model MR Is not feasible it is necessary to follow a blanch and bound procedure to search for the global optimum. This procedure requires a valid upper bound on the global optimum. This can be generated through heuristics or by solving directly the nonconvex model. For this purpose, the process network model is formulated in terms of the individual component flows and the nonconvex equations for the splitters are included. Equation (9) was also used in this work to model the splitters due to it is better numerical behavior. The solution to the model MR was used for the good initial point. In many instances, it was not possible to solve these nonlinear problems with MINOS 5.2. The nonlinear models were solved using CONOPT in GAMS 2.25.

During the branch and bound procedure a tree search is generated. Of the set of open nodes, these are the nodes that have a lower bound that is e-smaller than the current upper bound, the node with the smaller upper bound is selected to branch on. The splitter units are the units that are approximated, and of these, the splitter that has the largest difference

between its approximated and actual individual component flows is selected. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. The branching is done in the selected splitter over the concentration of the component that has the largest difference.

First, the actual concentrations for the individual component flaws in the LP solution (*) for the splitters are calculated by,

$$z_j^k = \frac{f_j}{F^k} \quad \text{for all the inlet streams to splitter} \qquad (26)$$

The splitter unit m is then selected according to the equation.

m = arg max$_k$ c mitt ($\sum_{S_k} \sum_{j=1}^{N}$ | ty-z^FH) , (27)

Equation (27) represents the total difference between the LP solution for the flows after the splitter and the actual value of these flows considering the concentrations before the splitter. Once the splitter has been selected, the component in that splitter that has the largest difference. J\ is selected by,

J--aigma^(JL"I$_{S_m}$ f/ - z»^{e} F* 11 (28)

The following branching constraints are then used;

$$x_j^m \le z_j^m \text{ and } x_j^m \ge z_j^m \qquad (29)$$

To improve the upper bound it might be necessary to solve additional nonconvex problems. These can be solved after a given number of nodes using the solution of the node with the smallest upper bound as the initial point. In this work if there was no significant change in the lower bound of the new nodes with respect to the lower bound of the parent node (< 1%) a new nonconvex problem was solved.

Example 1

Consider the following pooling and blending problem by Harveley (1978). Two streams that have components A and B are mixed in a initial mixer a then go through a splitter to obtain two streams than can be mixed with an additional stream (see Fig. 5a). Two different products can be obatined and there are constraints on the concentration of component A in these products. The objective function consists of niimizing the cost that is given by the total flow of the streams times the cost coefficients, c_t, given in Fig. 5.

This problem has two local solutions. One has an objective function 0 = -100 and consists of only producing product 2. The other local solution, that corresponds to the global optimum, has an objective of + « -400 and here only product 1 is produced.

Model MR is formulated for this problem and the initial lower bound is 0_L = -500. The nonlinear model. MF, is solved using the solution of model MR as the initial point and an upper bound of $ s -400 is obtained. Since there is a gap between the bounds of the global solution a partition is performed. There is only one splitter that needs to be approximated and since there axe only two components it is irrelevant which one is selected since the composition bounds are related (eg. $x_B^L = 1 \bullet x^{*u}$). The actual value of the composition of A in the solution of model MR is used as the branching point (x^* = 0.0166) to generate two new subproblexns. The first subproblcm (x_A £ 0.0166) has a lower bound of 4_L = -100 and the second subproblem (x^* £ 0.0166) has a solution of fa. = -400 (see Fig. Sb). Both of these bounds are greater or equal than the upper bound, therefore the global solution has been found (x_A = 0.01).

Example 2

The next example is a separation problem taken from Kocis and Grossmann (1987). The original problem has binary variables in the formulation and they have been fixed to 1 for this example (see Fig. 6).

*

The top stream of the flash unit has 85% of the inlet flow of A and the bottom stream has 80% of the inlet flow of component B. In the column. 97.5% of the inlet flow of A goes to the top whereas 95% of the inlet flow of B goes to the bottom stream. Hie total flow to the flash unit and the column have to be greater than 2.5 and smaller than 25, whereas the total flow of each of the two feed streams has to be less than 25. The objective function is given by,

$$\phi = 52 + 10 F! + 8 F_2 + F_4 + 4 F_5 - 35 Pj^A - 30 P_2^B \qquad (30)$$

The initial lower bound for this problem is 0_L = -513.22 and it is infeasible for the original NLP model. A nonconvex problem is solved using CONOPT with the solution of model MR as the initial point obtaining an upper bound of Φ = -511.87 and the relative gap is only 0.3%. Again only one splitter is present in the network and a partition can be performed using the concentration of component A in this splitter. The lower bounds for the new two subproblexns are *fa* = -511.87 (x_A £ 0.5121) and Φ_L = -511.80 ($x_A \geq$ 0.5121). Both solutions are greater or equal than the upper bound and the global solution has been obtained. In the global solution Fi = 8 and F_2 = 25, and 11% of the inlet flow lo the splitter is directed to the flash. 76% to the column and the rest bypassed to P_2.

Example 3

TIUs example corresponds to a separation problem with three feeds and three product streams. The network configuration and product specifications are given in Fig. 7. The objective function is given by

$$> « 4 S i + 1.5S2 + 4S3 \qquad (3D$$

The initial lower bound is 4_L = 138.18 and the nonconvex problem MP is solved obtaining a solution of $ = 138.7. The gap between these bounds is less than 0.4 %. The global optimum for this tolerance is shown in Fig. 8.

Sharp separation networks

In order to illustrate the application of the above algorithm to a specialized case where the structure can be further exploited, the problem that will be considered is the synthesis of separation networks with single feed and mixed products that consist of sharp separators and bypasses. It is assumed that a single feed with N components must be separated into K specified multicomponent product streams. The components are ordered from the lightest to the heaviest.

A modification of the superstructure proposed by Floudas (1987) for this problem is used (see Fig. (9)). The superestructure consist of N-l separators. Separator i performs the task of removing component number 1 to number! at the top of the separator and components number i+1 to N at the bottom of the separator. The feed to the network is split into N-l streams, F_t, that go to the separators and K streams, a_k. that bypass the network to go to the products. Each stream $F_{\{}$ is mixed before the separator i with streams that come from the top and bottom streams from the other separators to obtain the inlet stream to separator i, S^

The outlet streams of separator i are the top, $T_{(}$, and the bottom, B_t. These streams, T, and B_t. are each split into streams, PT_t^k and PB^ respectively, that go to the K products and into streams, KIV' and.RBf. that are redirected to the other separators. The top stream of separator i, T_{it} can be redirected only to the separators 1 to i-1 since it can only contain components number 1 to i. It would not be optimal to send part of this stream to any separator from i+1 to N since no separation would be achieved and a bypass of these separators would achieve the same separation with smaller flows. *KTf* is the flow redirected from the top of separator i to separator i\ In the same fashion that with the top stream, the bottom of separator i, B,, can be redirected only to separators i+1 to N-l since it can only

contain components that are separated by these sharp separators. RB/ is the redirected flow from the bottom stream of separator i to separator i\

Model

It will be assumed that the objective function can be expressed as a linear function that depends on the total flow to each separator. The model expressed in terms of concentrations and total flows has the following form:

min •= $\sum_{i=1}^{N-1} c_t S_i$			(32.1)
st. reeo!•» $\sum_{t»i}^{N-l}$ 2u *i * $\sum_{k*i}^{K}$ *m®k*			**IOZ.ZJ**
L_{ij} s F | L_j	**for all i and j**		**(32.3)**
Si» F| + $\sum_{t'«l}^{1-1}$ RB,^{1} + $\sum_{!'«*•!}^{N-l}$ X Kiy	for aU i		(32.4)
Si xsq = flj + $\sum^{1-1}$ RBf^{1} x^j + $\sum^{N-l}$ KT$_r^{!}$ xtrj	**for all i and j**		**(32.5)**
S i s ^ + Bi	**for alii**		**(32.61**
$T_i\, xt_{ij} = S_i\, xs_{ij}$	**for all i and ord(j) ≤ i**		**(32.7)**
$xt_{ij} = 0$	**for aU i and ord(j) > i**		**(32.8)**
$B_i\, xb_{ij} = S_i\, xs_{ij}$	**for all i and ord(j) > i**		**(32.9)**
$xb_{ij} = 0$	**for all i and ordQ) ^ i**		**(32.10)**
T_f *m* $\sum_{1*1}^{t-i}$ RTi^{r} + $\sum_{k«l}^{K}$ FT$_t^{k}$	for aU i		(32.11)
Bi s $\sum^{N-l}$ RB^^{f} + $\sum^{K}$ PB$_t^{k}$	**for all i**		**(32.12)**
$P^k = \sum_{i=1}^{N-l} PT_i^k + \sum_{i=1}^{N-l}$ PI3^ + 0^	Tor'alt k		(32.13)
P_{kj} = $\sum_{i*l}^{N-l}$ PT,^{k} xt$_{ij}$ + $\sum_{i»l}^{N-l}$ PB,^{k} *xbq*+ a_k z,	for all k and j		(32.14)
$\sum_{ord(j)=1}^{i} xt_{ij}$ = 1	for	alii	(32.15)
$\sum_{ord(j)=i+1}^{N}$ xb,j = 1	**for all i**		**(32.16)**
$\sum_{ord(j)=1}^{N}$ xs,j = l	**for alii**		**(32.17)**

Si, $T_{|t}$ B,. Fi. RT/. RBt*'. PT$_t^{k}$. PBi^{k}. c*. *t*^ xs,j, xty. $xb_{ij} \geq 0$

The parameters Feed, zj, P^k and p_{kj} represent the total feed, composition of the feed, total flow of product k and component flow of component j in product k, respectively. The variables xsq, xty and xbg are the concentrations of component j in the inlet stream to separator 1, top of separator 1 and bottom of separator i, respectively.

The objective function (32.1) is a linear function of the inlet flow to the separators. Equation (32.2) is the total mass balance in the inlet splitter and equation (32.3) is the component mass balance. Equations (32.4) and (32.5) are the total and component mass balances for the mixer i before the separator L The material balances for separator i are given by equations (32.6)* that is the total mass balance for the separator, equations (32.7) and (32.8) that are the component balances for the top stream and indicate that nothing from components number 1+1 to N-l is in the top of the separator, and equations (32.9) and (32.10) that are the component mass balances for the bottom streams. Equations (32.11) and (32.12) are the overall mass balances for the splitters of the top and bottom streams alter separator 1. The equations that state that the concentrations of the outlet streams should be the same that the inlet stream in a splitter have been already substituted. Finally, equations (32.13) and (32.14) are the overall and component mass balances for the mixer for product k.

Model (32) corresponds to a formulation of the type of model MX where the distribution coefficients are known and restricted to 0 or 1. Some simplifications have been made to avoid including many irrelevant variables (e.g. not to define concentrations for the streams that go the top i to product k). Although, some of the linear constraints in this formulation are redundant, they can become nonredundant in the linear relaxation as will be shown in Example 4.

Equations (32.5). (32.7). (32.9) and (32.14) involve nonconvex terms. This model can be reformulated as in model MR by introducing individual component flows and the linear equations (15M18) and (7) according to the approach illustrated earlier to obtain a model in the form of model MR The resulting reformulated model is as follows,

$$\min \phi = \sum_{i=1}^{N-1} c_i S_i \quad (33.1)$$

$$\text{st} \quad Feed = \sum_{i=1}^{N-1} F_i + \sum_{k}^{K} X_k \quad (33.2)$$

$$f_{ij} = F_i z_j \quad \text{for all i and j} \quad (33.3)$$

$$S_i = F_i + \sum_{t=1}^{i-1} RB_t^i + \sum_{t=i+1}^{N-1} RT_t^i \quad \text{for all i} \quad (33.4)$$

$$s_{ij} = f_{ij} + \sum_{t=1}^{i-1} rb_{tj}^i + \sum_{t=i+1}^{N-1} rt_{tj}^i \quad \text{for all i and j} \quad (33.5)$$

Sr-TV+'B, for alii (33.6)

t_n = sij for all i and ord(jJ £ i (33.7)

t_f * 0 for all i and ord(j) > i (33.8)

$b_{,jSS}$Sg for all i and ord(j) > i (33.9)

t,j = O for all i and ordfl) £ i (33.10)

T, = $\sum_{1*1}^{t-}$ sW + $\sum_{k«l}^{K}$ lPT$_4^k$ for all i (33.11)

B_i *m* $\sum_{i'=i+1}^{N-l}$ **£RB,^{if}** + $\sum_{k=1}^{K}$ £PB$_t^k$ for all i (33.12)

p* = $\sum_{i=1}^{N-1}$ **TFT$_t^{k''}$** + $\sum_{i=i}^{N-1}$ **£PB|^k** + otk for all k (33.13)

Pki * $\sum_{i=1}^{>M}$ 5jtt,^{k} + $\sum_{i=1}^{riti}$ Xpb^$^{k+}$ ctk z, for all k and j (33.14)

t« = $\sum_{i'»l}^{1*1}$ Z ^ / + $\sum_{k«l}^{K}$ Zptij^k foraUiandj (33.15)

b« = $\sum_{l'*t+l}^{N-l}$ Zrbij$^{4'}$ + $\sum_{k»l}^{K}$ £pb«^{k} for all i and j (33.16)

$\sum_{ord(j)=I}^{i}$ **xt$_{ij}$** = 1 for alii (33.17)

$\sum_{ord(j)=i+1}^{N}$ **xb$_{ij}$ = 1** for all i (33.18)

T_i = $\sum_{ord(j)=1}^{i}$ **t_{ij}** for alii (33.19)

B,s $\sum_{ord(j)=i+1}^{N}$ Ib« for alii (33.20)

PTi^k= $\sum_{oixi(j)»i'}^{i}$ Tpt<*^{k} for alii and k (33.21)

PB,^{k}= $\sum_{ord(j)=i+1}^{N}$ Ipbq^k for alii and k (33.22)

RT/ = $\sum_{ord(i)-l}^{i}$]DV for all i and i^f < i (33.23)

RB$_i^r$ = $\sum_{ord(j)=i+1}^{N}$ Irb/ for all i and r > i (33.24)

Equations (15-18) for t_{4J}, rt_{tJ}^l and pt_{tj}^k in terms of xt_{ij} and the total flow of its respective stream.

Equations (15-18) for bq, rbq* and pb_{fj}^k in tenns of xb_{ij} and the total flow of its respective stream.

S_{ij}. T_{lt} Bt. F_4. *KTf*, RB,$^{1'}$, PT,^{k}. PB^. a_k. f,j. $xs_{,jf}$ $xt4_{jf}$ xb,j ^ 0

s_{ij}, t_{ij}, b_{ij} pt_{ij}^k, pbj ^{k} . rt,/. rby^1* ≥ 0

It is not necessary to include equations (15)-(18) for the inlet component flows to the separator, Sq. since the variables xsy only appear in these equations. Also, the component flows, S|j9 only appear in mixers and sharp separators units which can be exactly represented in terms of the individual component flow equations (33.5). (33.7) and (33.9). Equations (33.15)-(33,16) that are the component mass balances for the splitters of top and bottom streams have been included accordingly to the reformulation previously presented. Equations (33.19M33.24) relate the total flow and the individual component flows for the splitter streams.

Preprocessing

The proposed Superstructure (Fig. 9) allows to bypass certain amount of the feed to the product k. Ok, without having to go through the separation network. The amount of the product k that is not bypassed has to be processed In the separation network and it will be denoted as the 'residual product*. Hence, the total 'residual product k^f is given by $(P^k - a_k)$ and has the component flows given by (p^ - o^Zj)(see Fig. 10).

The global optimal solution of model (32) is a network in which all the 'residual products[1], have at least one component with a zero flow. The reason that it is not optimal to separate a stream in the network and later to remix it. The same degree of separation can be achieved using a bypass that does not incur any cost in the objective function.

Consider the second separator in the solution obtained by Floudas (1987) to his second example (see Fig. 11). For this subnetwork of the complete structure the "upper 'residual product' has components B and C present. The components are being separated and remixed again. The same outlet flows can be obtained with a smaller input flow to the separator as it is shown in Fig. 11. Note that both 'residual products' have components with zero flow.

It should be clear that if there was not a component with zero flow in the 'residual product[1], then there is part of this stream that could have been obtained by just bypassing the network. This in turn does not incur in any cost, whereas going through the network has a positive cost. The above condition gives a lower bound for the bypass to each product. This also corresponds to the largest amount that can be bypassed since all this flows in the 'residual flow[1] have to be positive. In this form the bypass can be precalculated without affecting the global optimality of the solution.

The bypass to product k is given by the maximum amount that can be sent to product k without having a negative flow; that is.

$$a^* = \min_j t ^4 \qquad (34)$$

where z_j is the feed composition and p_{kj} is the flow of component j in product k. The component flows for 'residual product' k, γ_{kj} are given by.

$$\gamma_{kj} = p_{kj} - \alpha_k z_j \tag{35}$$

Key component bounds

Wehe and Westerberg (1987) proposed using lower bounds for the flow of the key components in separator i. These bounds are based on the fact that separator i is the only unit that can perform the task of separating component number i from component nutaber i+1. They are redundant for the nonlinear formulation (32) but they are relevant for the linear relaxation in (33). To calculate them, it is necessary to determine in each product what is the difference between the two key components of separator 1 with respect to the concentrations in the feed. The lower bounds in separator i* for the flow of the key components in the top (component j1) and bottom (component j2) streams are given by:

$$t_{ij1} \geq \sum_{k=1}^{K} \{ \gamma_{kj1} - z_{j1} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \tag{36}$$

$$b_{ij2} \geq \sum_{k=1}^{K} \{ \gamma_{kj2} - z_{j2} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \tag{37}$$

where γ_{kj} is given by (35). It is important to include both bounds in the relax model (33) since there is no guarantee that the inlet flow to separator i has the same proportion between the key components as the feed. It is not known in which part of the sequence separator 1 will be placed, and it can be after a splitter that is not being approximated correctly.

The bounds in (36) and (37) can be extended to separation of components that are not adjacent in the feed. Consider component number i and component number i+3. There are three separators that can perform this task, separators i. i+1 and i+2. Cuts of the following form can be obtained,

$$t_{i\,i} + t_{i+1\,i} + t_{i+2\,i} \geq \sum_{k=1}^{K} \{ \gamma_{ki} - z_i \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \tag{38}$$

$$b_{i\,i} + b_{i+1\,i} + b_{i+2\,i} \geq \sum_{k=1}^{K} \{ \gamma_{ki+3} - z_{i+3} \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \tag{39}$$

Equations as the ones in (38) and (39) can be redundant compared to equations (36) and (37) and it is possible to detect this before solving the problem.

Relative flowrate constraints

These constraints are used when the relation between the flowrates of two components is known. In particular, consider component A in the last column of the network (we Fig. 12). None of the redirected streams contains component A. Therefore, the relative flowrate of component A with respect to the other components in the top stream has to be smaller than in the feed. This relation should remain valid after the top stream is split to the products and redirected flows.

In the separator previous to the last one, N-2, all the streams do not have component A except the one coming from the top of the last column. For this one it is already known that the relative flow of component A with respect to the other components is smaller than in the feed. This type of analysis can be done for component A and component N in all the columns yielding th$ following linear constraints for the splitters.

for alii and k

$ZAPtij^{k}$-ZjPt«A^{k}^O   forj *Aandord(j)≤i   (40)

z_N pb,j^{k} - z, pt*$_N^{k}$ £ 0   for ord(j) * N and ord(j) > i   (41)

foralliandi'>i

ZArt|j^{r}•ZjrtiA$^{4'}$^0 ^{for}J * ^{Aandord}W ^ *   W2)

for alii and f < i

z_N rb$_{ij}^{r}$ - z_j rb$_{iN}^{r}$ ≥ O   forord(J)*Wandord(j)>i   I43J

Boundson concentrationsand total flows

The approximations (15-18) require bounds for the total flows and component concentrations in the splitters. The lower bound for the total flow of the top and bottom streams is given by the lower bound of the key components obtained in equations (36) and (37). For the outlet streams of the splittters, that are the redirected streams and the streams that go to the products, the lower bound is zero. The upper bound for the total flow of the top and bottom streams is given by the feed to the network of the components that can be present in each stream .that is,

T_t^{u}= Z(Feed-Xak)z, (upper limit t, lower limit ord(j)=1, k)   **for ail i**   **(44)**

B|«= ItFeed-IOfcJz, (upper limit N, lower limit ord(j)=i+1, k)   **forallj**   **(45)**

The upper bound for the streams after the splitter are given by.

$RT_i^{i'u} = T_i^u$ for ail i and i' < i (46)

$RB_i^{i'u} = B_i^u$ for all i and i'> i (47)

$PT_i^k = \sum_{ord(j)=1}^{i} \gamma_{kj}$ for all i and *k* (48)

$PB_i^k = \sum_{ord(j)=i+1}^{N} \gamma_{kj}$ for alii and k (49)

The lower bounds for the concentrations are zero except for the key components in the separator for which the lower bounds are given by the lower bound of its flow divided by the upper bound of the total flow of that stream. The upper bounds in the concentrations are given by one minus the lower bounds of the other components.

The solution of the linear programming model (33) provides a lower bound to the global optimum since this model is a valid relaxation of the nonconvex model (32). This lower bound is obtained by solving the LP model for the 'residual products' in 135) with the addition of the valid constraints (36)-(43).

The upper bounds are generated solving mode} (32) in terms of the individual flows for the 'residual products'. When additional nonconvex problems are solved to improve the current upper bound it can happen that very similar initial point are generated. In this case, a new nonconvex NLP is solved in which bounds over the total inlet flows to the separators (SJ are included. For this purpose the values of these variables in the LP solution (S_4*) are used such that the current incumbent solution is no longer feasible.

Example4

Consider the 3 component example proposed by Floudas and Agganval (1990). An equimolar feed has to be separated into two products as show in Fig. 13. The objective function is given by

$$\Phi = 0.2395 + 0.00432 S, + 0.7584 + 0.01517 \&* \quad (50)$$

The bypass to products 1 and 2 can be calculated according to equation (34) and the 'residual product' component flows are obtained through equation (35) (see Fig. 14). The total bypass to product 1 is a, = 90 and the bypass to product 2 is ct_2 = ISO and the feed has a concentration of z_A = 1/3, z_Q = 1/3 and Zc = 1/3. In this form the 'residual product' 1 is YIA = 0, YiB = 20 and y_{lc} = 0 and the residual product' 2 is Y_2A = 20, Y_{2B} = 0 and Y2C = 20. Additionally, lower bounds on the flow of the key components in both separators are obtained

using equations (35)-(36). The key components in separator 1 are component A in the top and its flow has to be at least 20 and component B in the bottom has to have at least a flow of 20. In the top stream of the second separator at least 20 units of component B have to be separated from 20 units of component C in the bottom stream. It is important to note that after preprocessing the network several suboptimal solutions have been cut off. One of these suboptimal solutions for this particular data is a parallel configuration of both separators (there are situations in which a parallel configuration can correspond to the global solution as will be shown in example 5). In this example the direct or indirect sequence have a lower objective function. Both of these configurations are local solutions with an objective function value of 4 » 1.8639 for the direct sequence and 4 = 2.081 for the indirect one. In some instances, MINOS 5.2 had problems converging even in this small example.

The LP (33) is formulated for this problem, giving a lower bound of (^ = 1.8639. The approximations are exact and therefore this solution is a feasible solution of model (32) proving that it corresponds to the global optimum. Hence, convergence is achieved in one iteration. The optimum solution corresponds to the direct sequence shown in Fig. 15. It should be noted that if the linear mass balances for the mixer for product 2 were not considered since they are redundant for the nonlinear formulation, a lower bound in the relaxed model of Φ_L = 1.12 is obtained, this shows that it is relevant to include all the mass balances in the linear model in order to tighten the lower bound.

Example 5

In the approach proposed by Wehe and Westerberg (1987) for the case of 3 components only the direct and indirect sequences are considered and both options can be modelled as LP problems since no mixing is required for these separation networks. However, this example shows that parallel configurations can be also globally optimal and that they are not excluded by the method proposed in this paper. To be able to consider parallel configurations or any combination of parallel with direct or indirect sequences it is necessary to model a superstructure in which mixing is allowed (like in the structure used in Fig. 13). Here, nonconvexities arise in the mass balance equations after the separators.

Consider that an equimolar feed is to be separated into the two different products given in Fig. 16. The objective function is to minimize the sum of the total flows into the separators. The same procedure that in the previous example is followed and the bypass can be precalculated by equation (34). The solution to the model (32) yields Φ_L = 12 and since it is a feasible solution of model (32) it corresponds to the global optimum (see Fig. 16). Note that the solutions for the direct or indirect sequences have an objective function of Φ =16.

Branch and Bound

If there is a gap between the lower and upper bound a branch and bound search is performed. It is only necessary to do the search over the variables involved in the nonconvex terms. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter* In this way. it is necessary to check the approximation for the concentrations in the splitters of the top and bottom streains of the separator. Equations (26)-(29) for the splitters of top and bottom streams are used to perfom the branch and bound search.

Results

Table 1 summarizes the results of the earlier examples 1 to 3 and of the sharp separation network examples 4 to 12. The number of variables is the total number of variables that are included in the reformulated and relaxed model (33) for that example. The lower bound is the initial bound that *fs* obtained by solving model (33) over the entire feasible space. The inital gap represents the percentage difference between the initial lower and upper bounds. When there is a zero initial gap it means that the first relaxed solution is feasible in the original problem thereby corresponding to the global solution. The column for nodes gives the total number of subproblems that where solved before converging to the global solution. A relative tolerance of 0.01 was used, except for example 2 were exact convergence was obtained after branching and example 12 for which a tolerance of 0.02 was used. It is important to note that the initial lower bound is tight and that it corresponds to a good estimation of the global solution. The largest differences are for example 1 with a 25% of difference and for example 12 with a 7% difference. The LP time refers to the time used to solve each relaxed model and the NLP time is the time used for solving a nonconvex model. It is possible to do updates using the previous LP solution and in this form have a more efficient implementation. The times are in seconds and the problems were solved on an IBM RS600/530 using GAMS 2.25 (Brooke et al. (1988)). MINOS 5.2 was used to solve the LP problems and CONOPT for the nonconvex NLP problems. A brief description of the example problems 6 to 12 is given below. It includes the specific data for the problem, the objective function and the topology of the network that is the global solution.

Example 6

This example corresponds to example 2 from Floudas (1987). In this case a linear objective function with the same cost coefficients is used and it is given by,

$$\Phi = 2.5\ S_1 + 3.0\ S_2 + 1.5\ S_3 \qquad (51)$$

The data for the composition of the products is given in Table 2.

The initial lower bound is Φ_L = 54.25 and an upper bound of 0 = 55.5 is obtained by solving the nonconvex problem. A partition of the feasible region is performed using the composition of component D in the bottom stream of separator 1. The first subproblem (x £ 0.166) yields a lower bound of Φ_L = 55.45 and the second subproblem (x ≤ 0.166) has a solution of Φ_L= 55.8. The latter is greater than the upper bound and the former is less than 1% of the global solution (see Fig. 17).

Exajnple7

This example is taken from Floudas (1987). The data for this problem is given in Table 3 and the linear objective function is given by:

$$\Phi = 2.5S_1+3.0S_2+L2S3 \quad (52)$$

The initial lower bound is Φ_L = 32.7 and it provides a feasible solution to the nonconvex problem. In this form the global solution (see Fig. 18) is obtained in one iteration. It is interesting to see that this solution also provides a better objective function for the concave objective function used by Floudas (1987) ($ = 10.65 versus $ = 13.68 which is 28% higher)

Example 8

Tills four component problem is taken from Wehe and Westerberg (1987). The data for the products is given in Table 4 and the objective function has the following form:

$$\Phi = 5.0 + 0.5\ S_1 + 4.0 + 0.3\ S_2 + 6.0 + 0.7\ S_3 \quad (53)$$

The first relaxed subproblem has a solution of 0_L =26.76 and it is infeasible for the nonconvex problem. A nonconvex problem is solved using CONOPT with the LP solution as the initial point. An upper bound of 4> = 26.79 Is obtained corresponding to the global solution (see Fig. 19) within a 0.1%.

Example 9

This example corresponds to example 1 from Wehe and Westerberg (1987). Table 5 provides the data for the product flows and the objective function is given by:

$$\Phi = 5.0 + 0.5\ S_1 + 9.0 + 1.0\ S_2 + 3.0 + 0.4\ S_3 + 6.0 + 0.6\ S_4 \quad (54)$$

A initial lower bound of Φ_L = 85.16 is obtained and the upper bound Is $ « 85.65. The difference is 0,5% and the global solution (see Fig. 20) is obtained in one iteration.

Example 10

This problem is taken from Floudas (1987) and the data is given in Table 6. The objective function is given by,

$$4 \text{ « } 1.2 \text{ Sj} + 3.0 \text{ Sj} + 2.5 S_3 + 1.5 S_4 \quad (55)$$

The JrHui lower bound is <^ = 156.56 and the upper bound is $ = 179.08. After 5 nodes the global solution of $ = 159.48 (see Fig. 21) is obtained.

Example 11

The data for this 6 component problem are given in Table 7 and the objective function has the following form:

$$4 \text{ »i.5 S, } + 3.0 \text{ Sj} + 2.0 \text{ S3} + \text{I.OS4} + 4.0 S_5 \quad . \quad (56)$$

The initial lower bound is 0_L = 173 and the upper bound is 4 » 179.11. After five nodes the global solution is obtained (see Fig. 22).

Example 12

This is a 6 component 4 products problem and the data are given in Table 8. The objective function is:

$$\$ \, m \, 5.0 \text{ Si} + 3.0 \text{ S2} + 2.0 S_3 + 2.5 S_4 + 4.0 S_5 \quad (57)$$

The initial lower bound is ϕ_L = 362 and the initial upper bound is 0 = 415.6. The global solution of 4> = 388 (with a 2% optimality gap) is obtained after 33 nodes (see Fig. 23).

Conclusions

A general procedure for the global optimization of process networks with multicomponent streams has been proposed. The basic idea relies on a relaxed LP model that is obtained through reformulation-linearization techniques that establish a clear relation between the component flow and the composition models for mass balances. The reformulated model combines both of these providing tighter lower bounds than other relaxations proposed in the previous work. The relaxed linear model has been embedded in a branch and bound procedure to obtain the global optimal solution.

As has also been shown, the algorithm can be further specialized to take advantage of the particular structure of sharp separation networks with single feed and mixed products. Here, it is possible to preprocess the problem to reduce the space over which the search is

conducted. The bounds that are necessary for the estimator functions in the relaxed model can be obtained without having to solve any subproblems. Different types of linear approximations that are nonredundant to the relaxed model are included to obtain a tighter lower bound.

Twelve examples for both general process networks and for sharp separation networks have been presented to illustrate the performance of the algorithm. As has been shown, only a small number of nodes are commonly needed in the branch and bound search to identify the global or e-global solution. Moreover, in many cases the initial lower bound is either the exact solution or a very good approximation to the global solution.

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Appendix A. Reformulation-Linearization to obtain the nonlinear constraints in model MF

The nonlinear equations in model MF. that can be expressed either as (8) or (9), can also be generated from model MX. For this purpose take the concentration model MX and consider equation (5),

$$x_j^k = x_j^l \qquad \text{(A.1)}$$

multiply by the valid bound constraint Xj^k £ 0

xfvfm^xf **(A.2)**

Use equation (5) for component J\

*xfxf**xtXf*^{k} **(A.3)**

Multiply by the valid bound constraints $F^k \geq 0$ and $F^1 \geq 0$,

$F^k x,^k F^l Xf^4 = F*\wedge^l F^k x\wedge^k$ (A.4)

that it is linearized to yield.

$$f_j^k f_j^l = f_j^l f_j^k \qquad \text{(A.5)}$$

which is precisely equation (8) for the splitters in the individual flow model MF.

Consider again equation (5),

$x,^k = \wedge$ IA.6)

multiply by the valid bound constraints $F^k \geq 0$ and $F^1 \geq 0$,

F^xfF^F'xj'F^{1}' IA.7)

that can be linearized to yield.

$$f_j^k F^l = f_j^l F^k \qquad \text{(A.8)}$$

Define the split fraction £ to be,

? » £ (A.9)

Equation (A.8) can then be expressed as

fjt^tff (A. 10)

which corresponds to equation (9).

Hence, the nonlinear equations (8) and (9) are redundant to model MX. Their linear approximations in general are also redundant in the linear reformulated model MR. Consider equation (A. 10), similarly to (15) one of its linear approximations is given by.

$$f_j^l \geq \xi^{lL} f_j^k + \xi^l f_j^{kL} - \xi^{lL} f_j^{kL} \qquad \text{(A.11)}$$

If there are no particular restrictions in the splitters, then the bounds for the split fraction variable are 0 £ $£^f \leq 1$ and using them in (A. 11) yields.

$$f_j^1 \geq f_j^{kL} \xi^1 \qquad \text{(A.12)}$$

The bound for the individual component flow is given by $f_j^{kL} = x_j^{kL} F^{kL}$; also $x_j^k = x_j^1$ and $\xi^f = \frac{F^1}{F^f}$ which leads to,

$$f_j^1 \geq x_j^{1L} F^1 \frac{F^{kL}}{F^k} \qquad \text{(A. 13)}$$

The estimator (15) for the same conditions ($F^{1L} = 0$) is given by

$$f_j^1 \geq x_j^{1L} F^1 \qquad \text{(A.14)}$$

Since the factor $\frac{F^{kL}}{F^k}$ is always less or equal than 1. equation (A. 13) is redundant. A similar analisis can be performed for the other estimators. Only when more specific bounds over the split fractions or the individual component flows are known, will these additional estimators be non redundant.

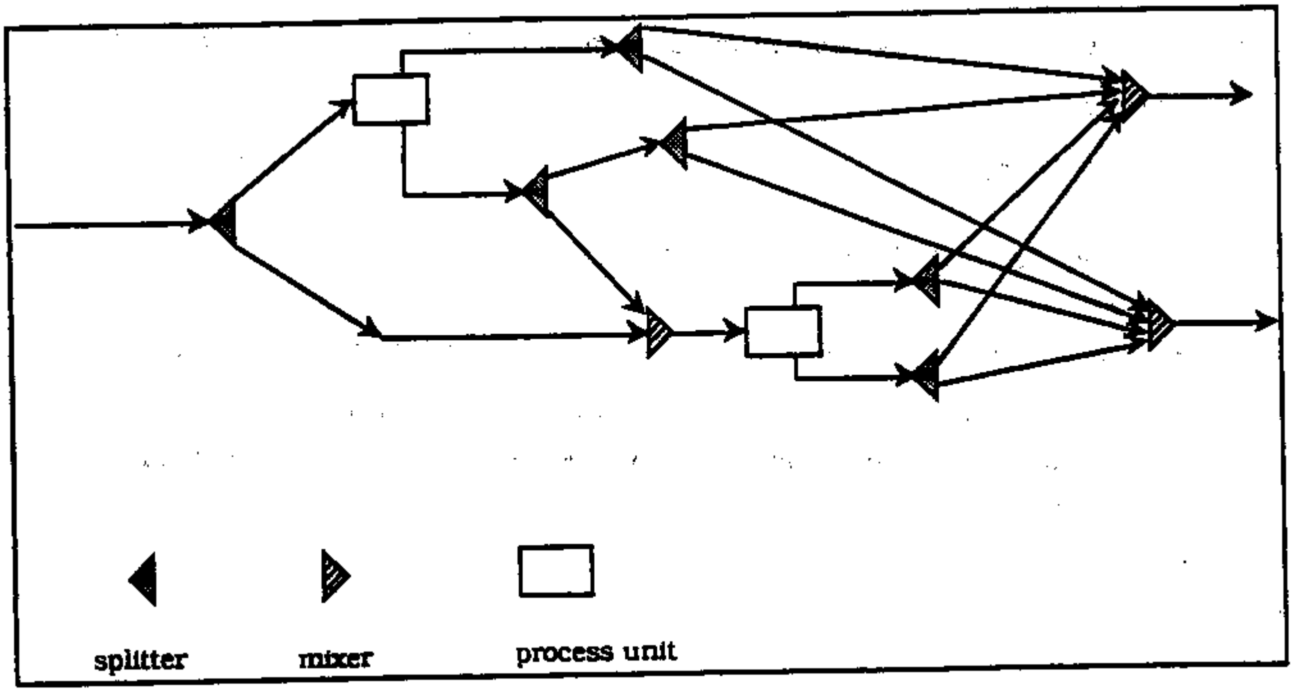

Figure 1. Process network with units, splitters and mixers.

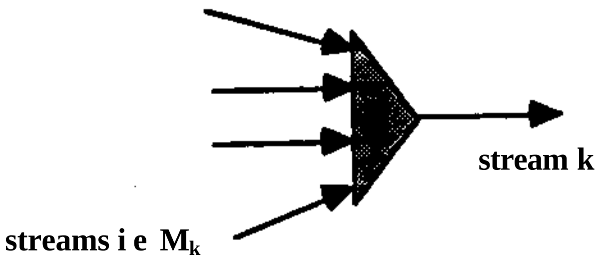

Figure 2. Mixer module.

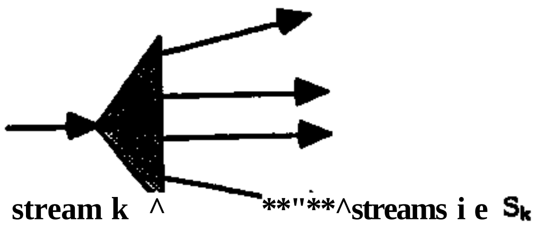

Figure 3. Splitter module.

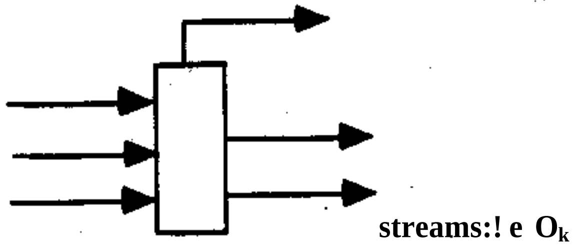

Figure 4. Process unit module.

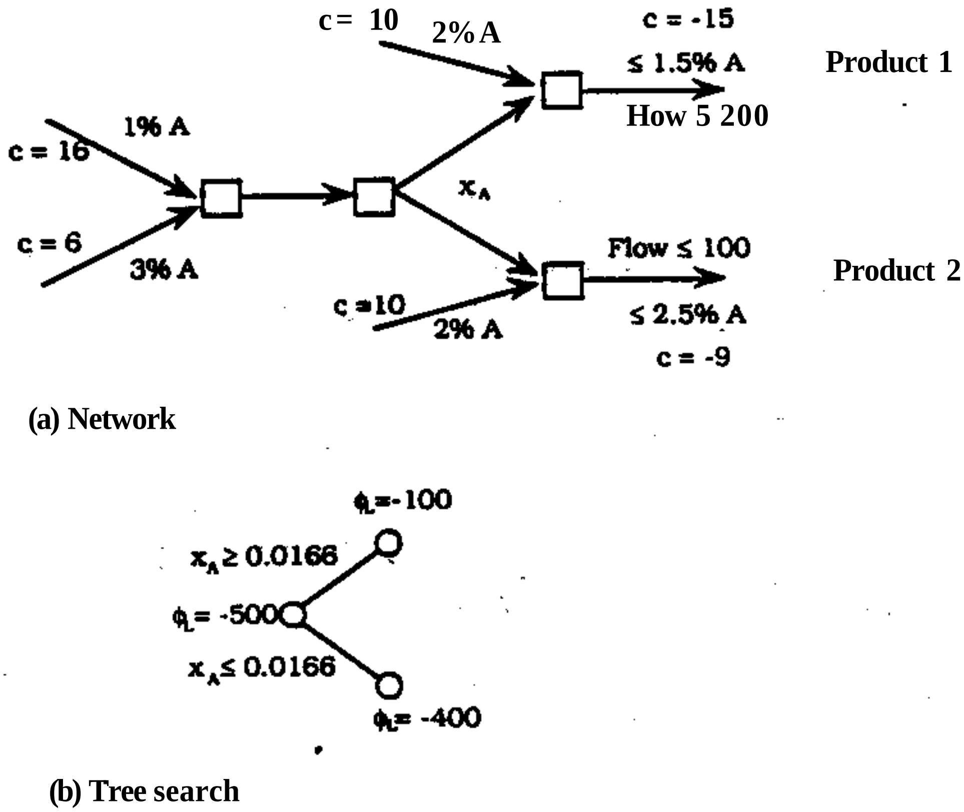

Figure 5. Network and branch and bound search for example 1

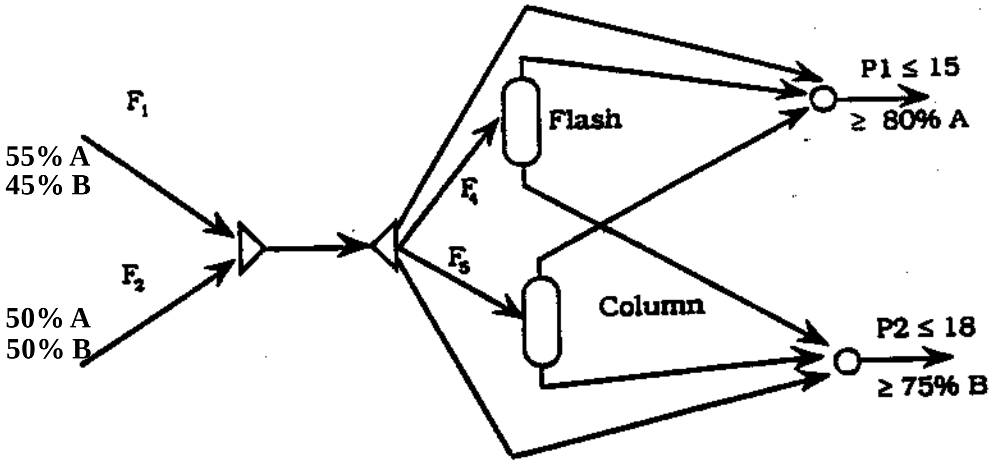

Figure 6. Network for example 2.

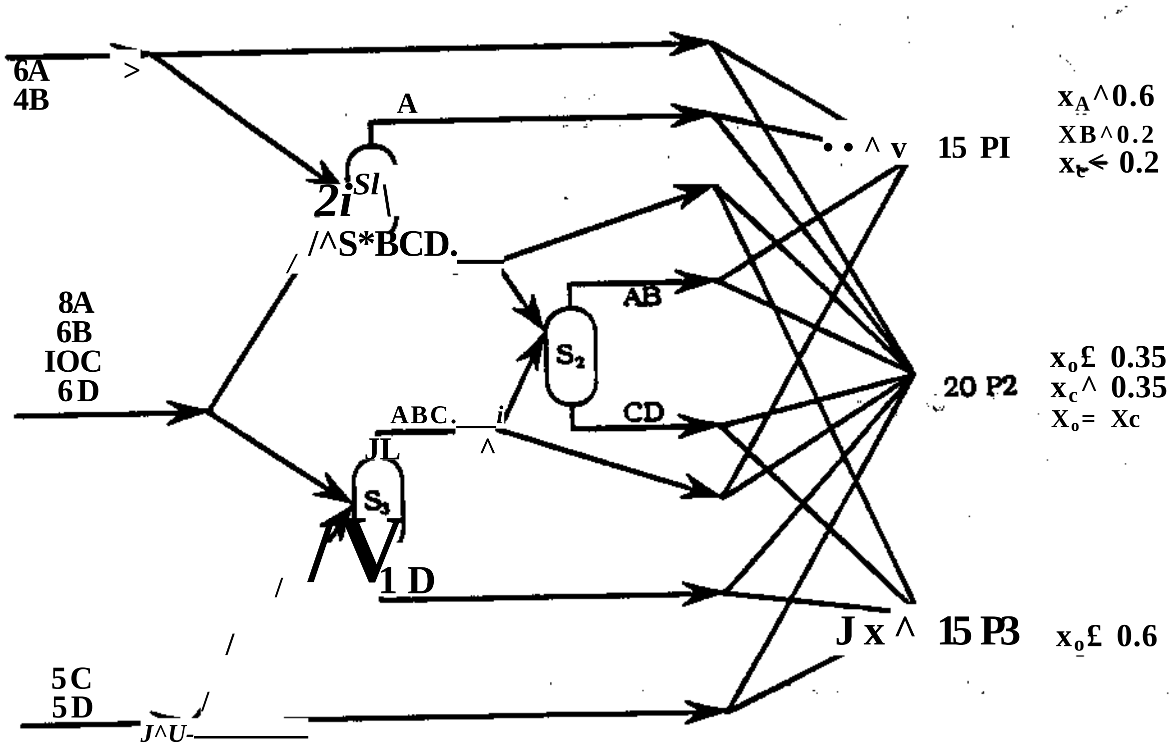

Figure 7. Network for example 3.

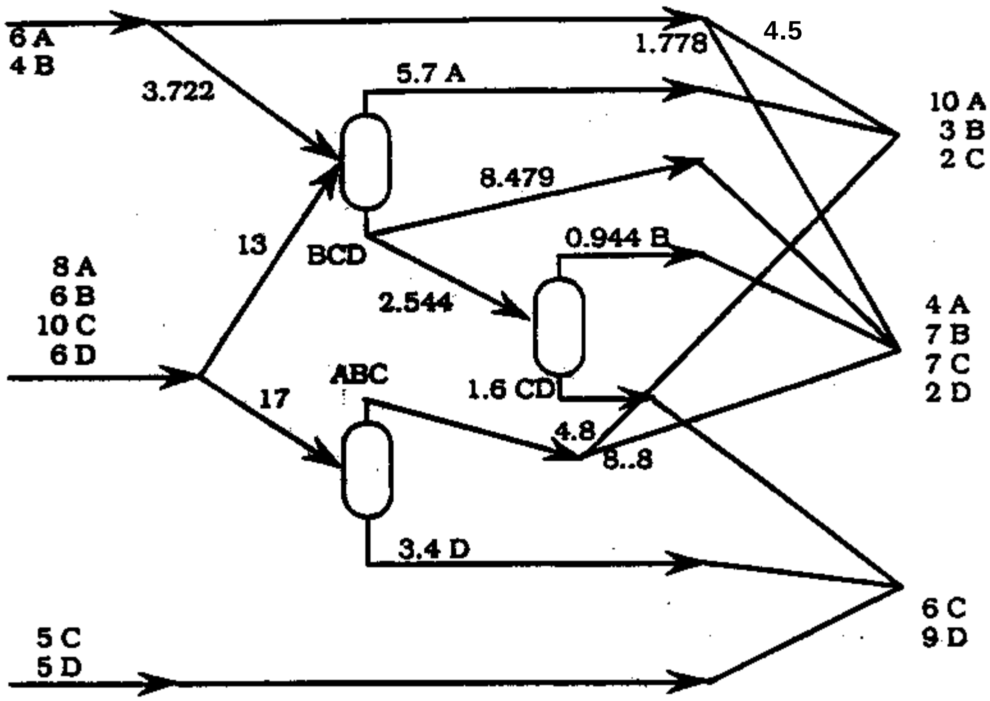

Figure 8. Optimal network for example 3.

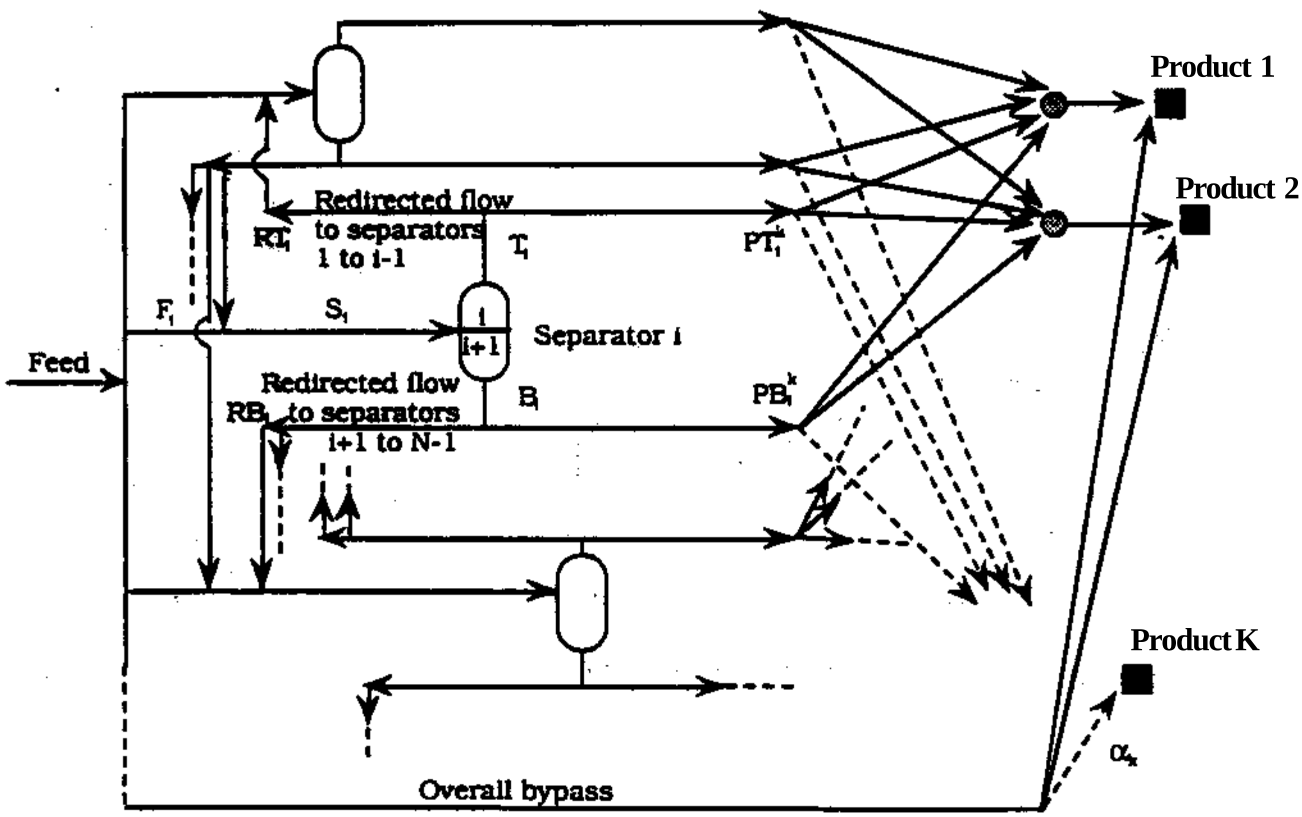

Figure 9. Superstructure for separation with sharp splits and mixed products.

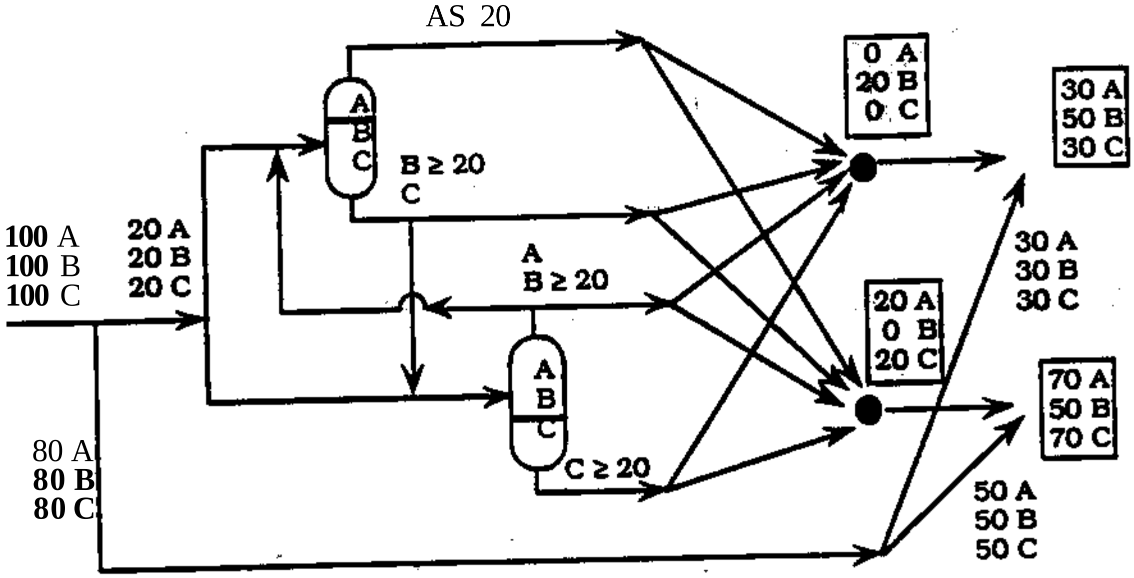

Figure 14. Residual products and key component bounds in example 4.

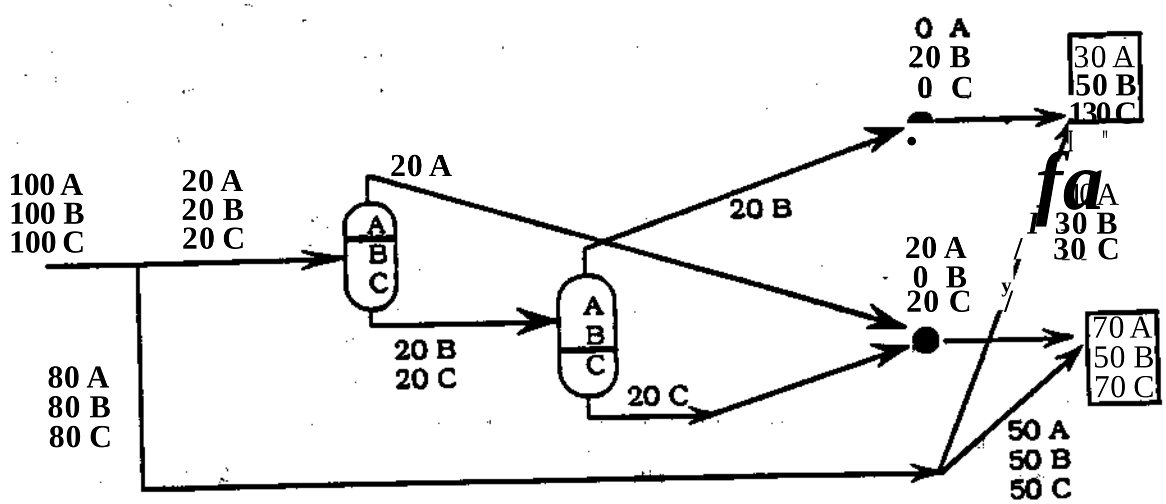

Figure 15- **Global** optimum solution of example 4.

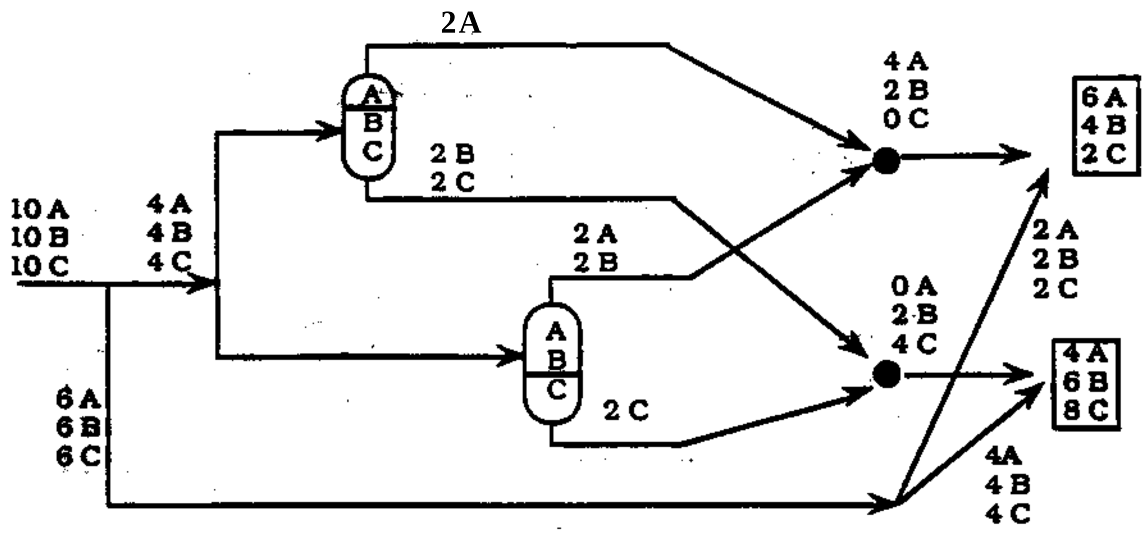

Figure 16. Global optimum solution of example 5.

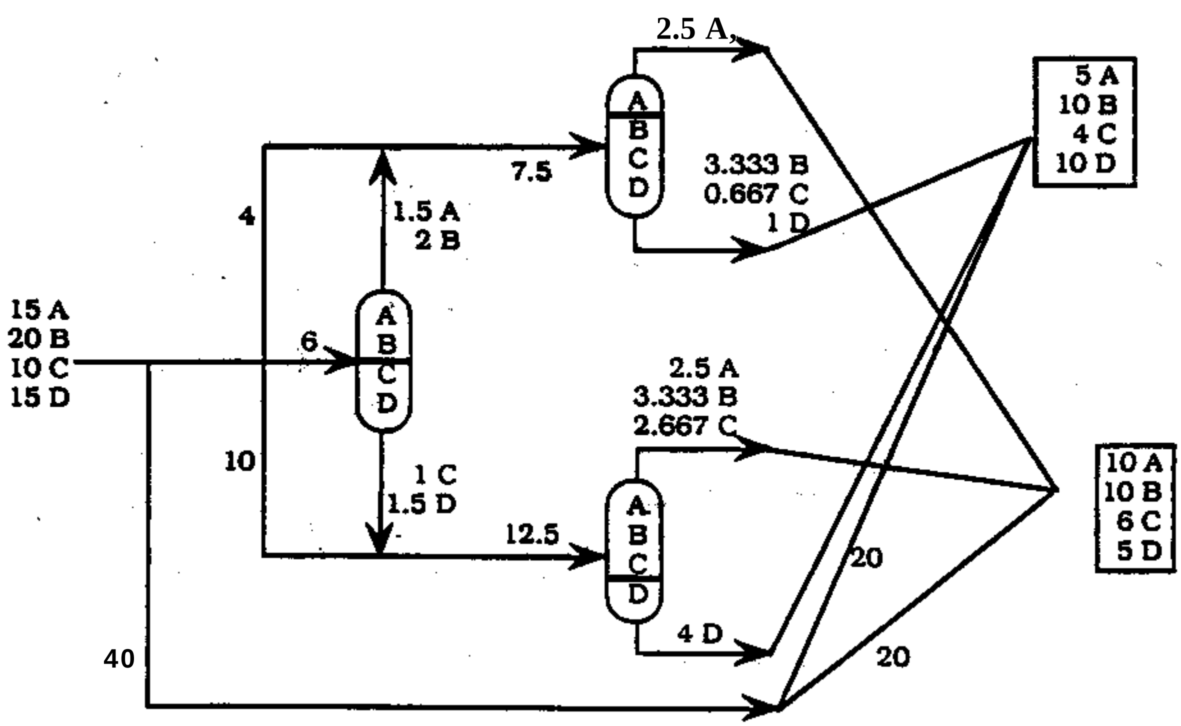

Figure 17. Solution of example 6.

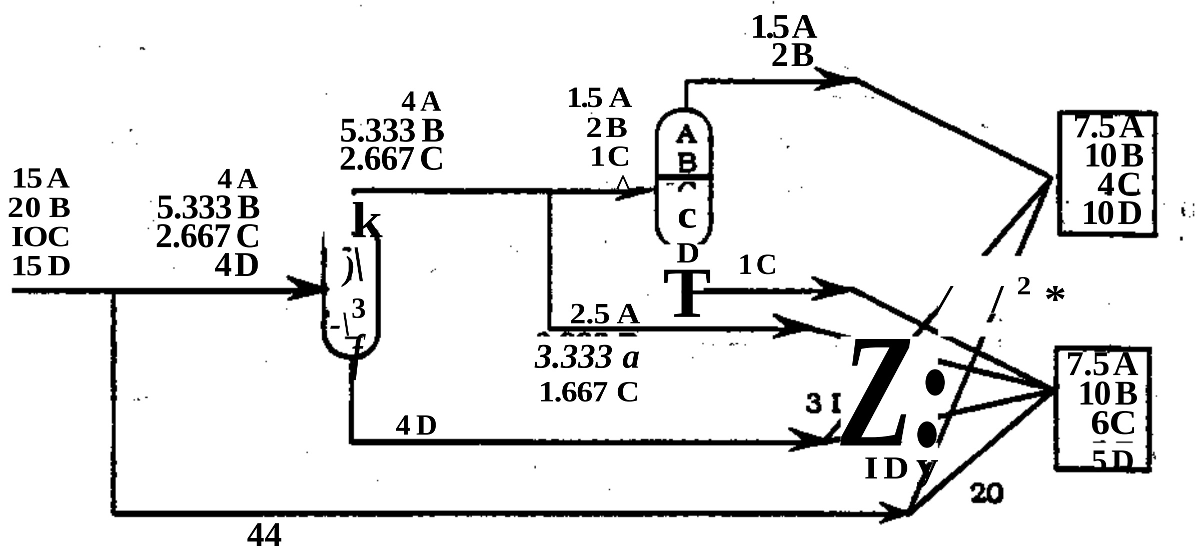

Figure 18. Solution of example 7.

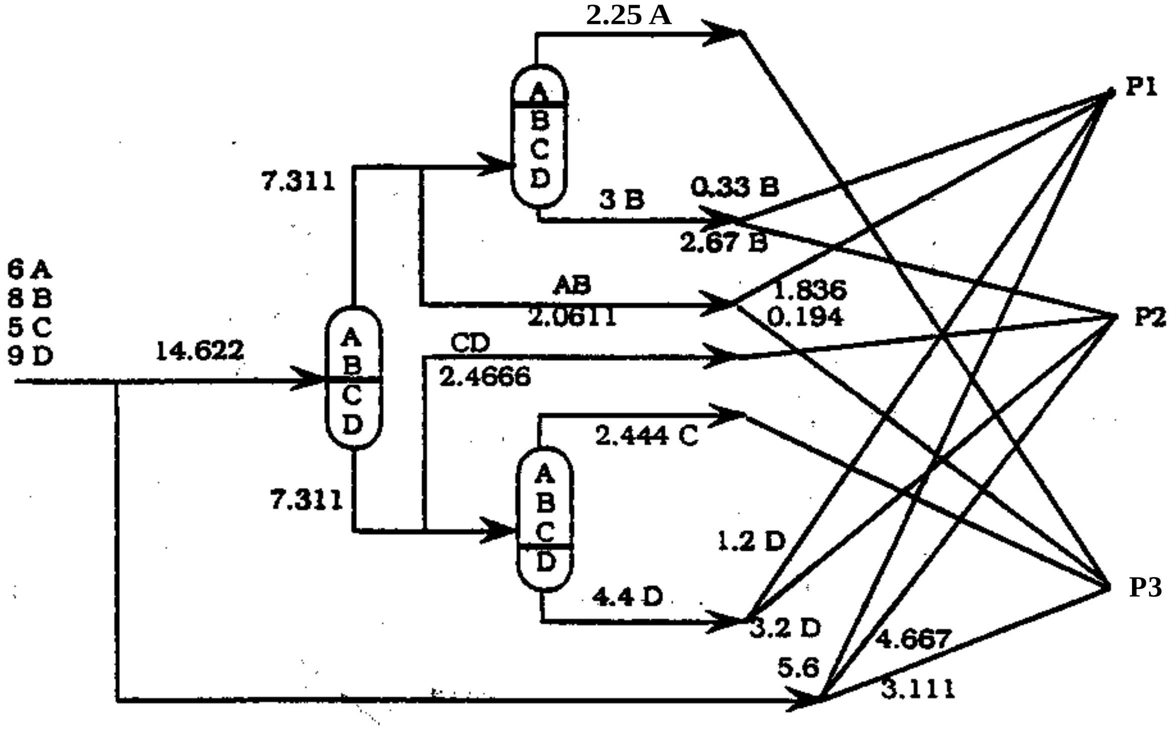

Figure 19. Solution of example 8.

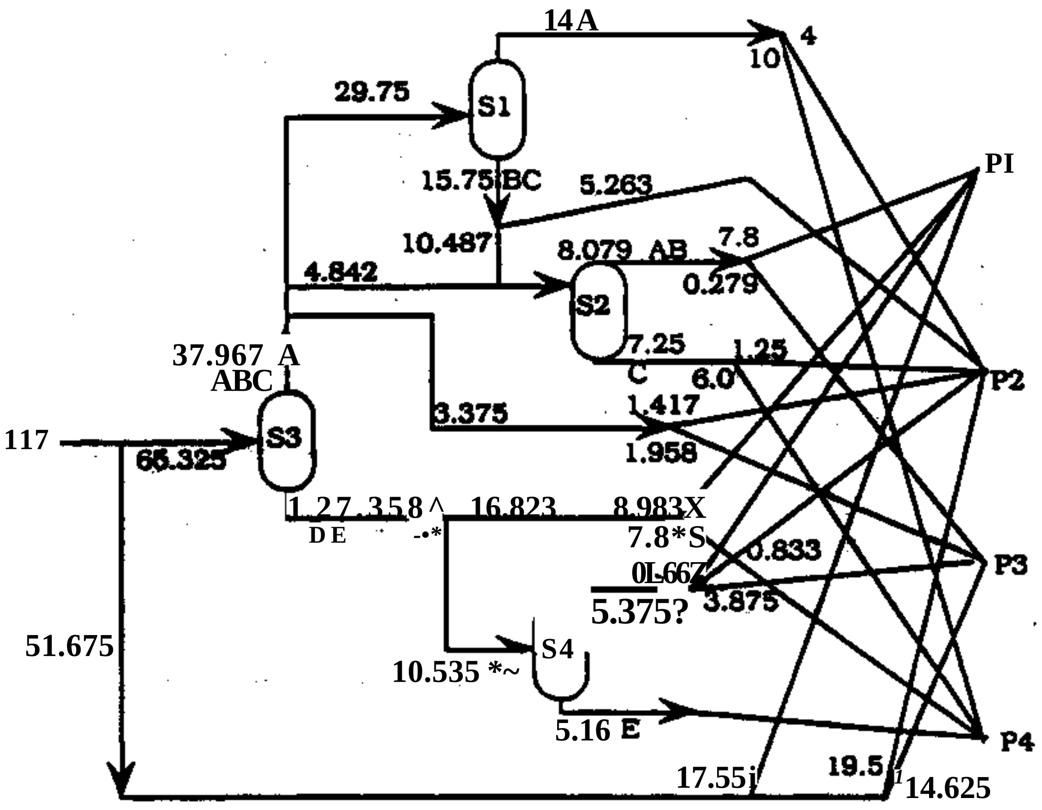

Figure 20. Solution of example 9.

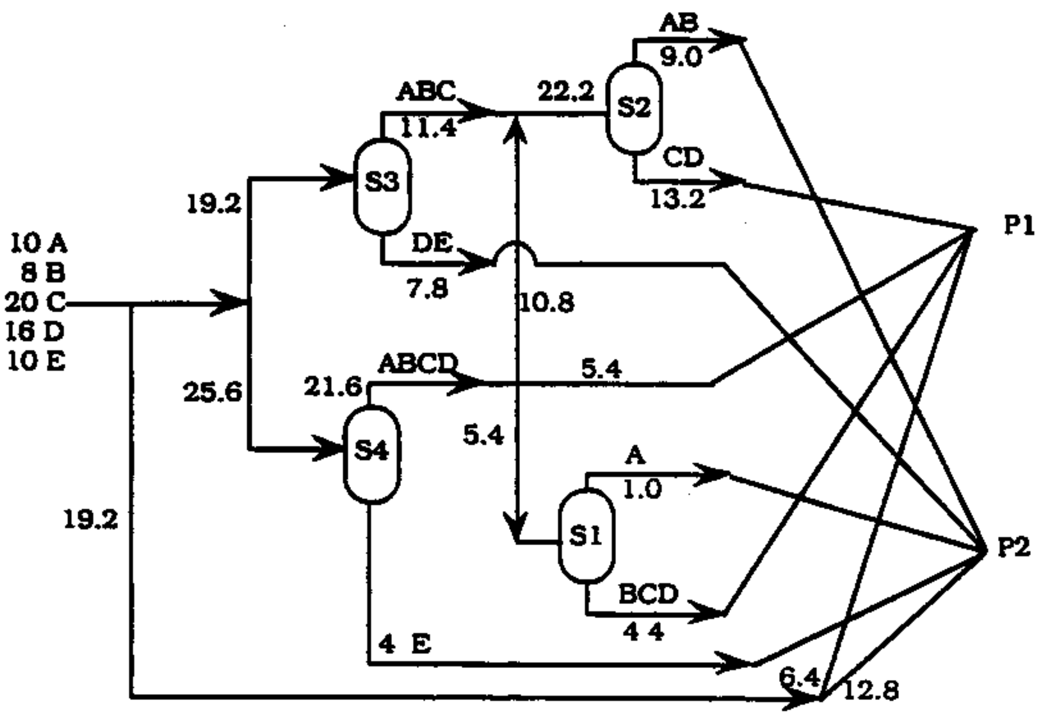

Figure 21. Solution of example 10.

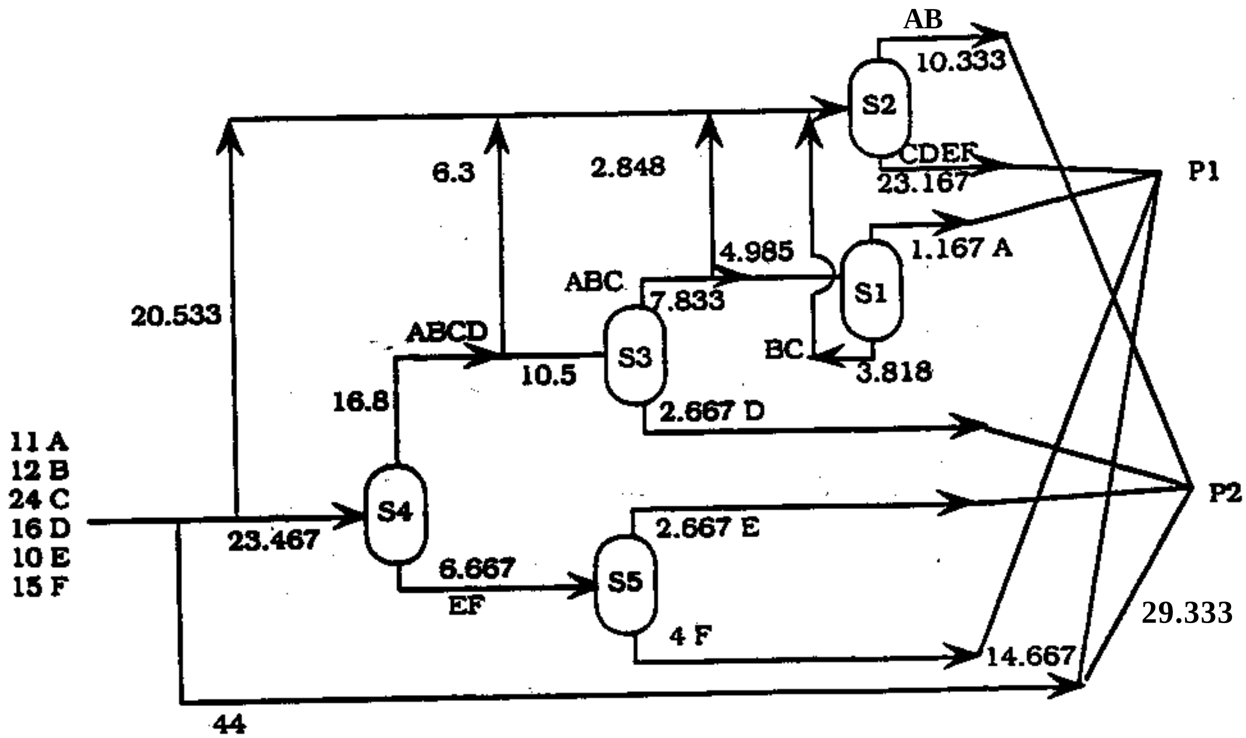

Figure 22. Solution of example 11.

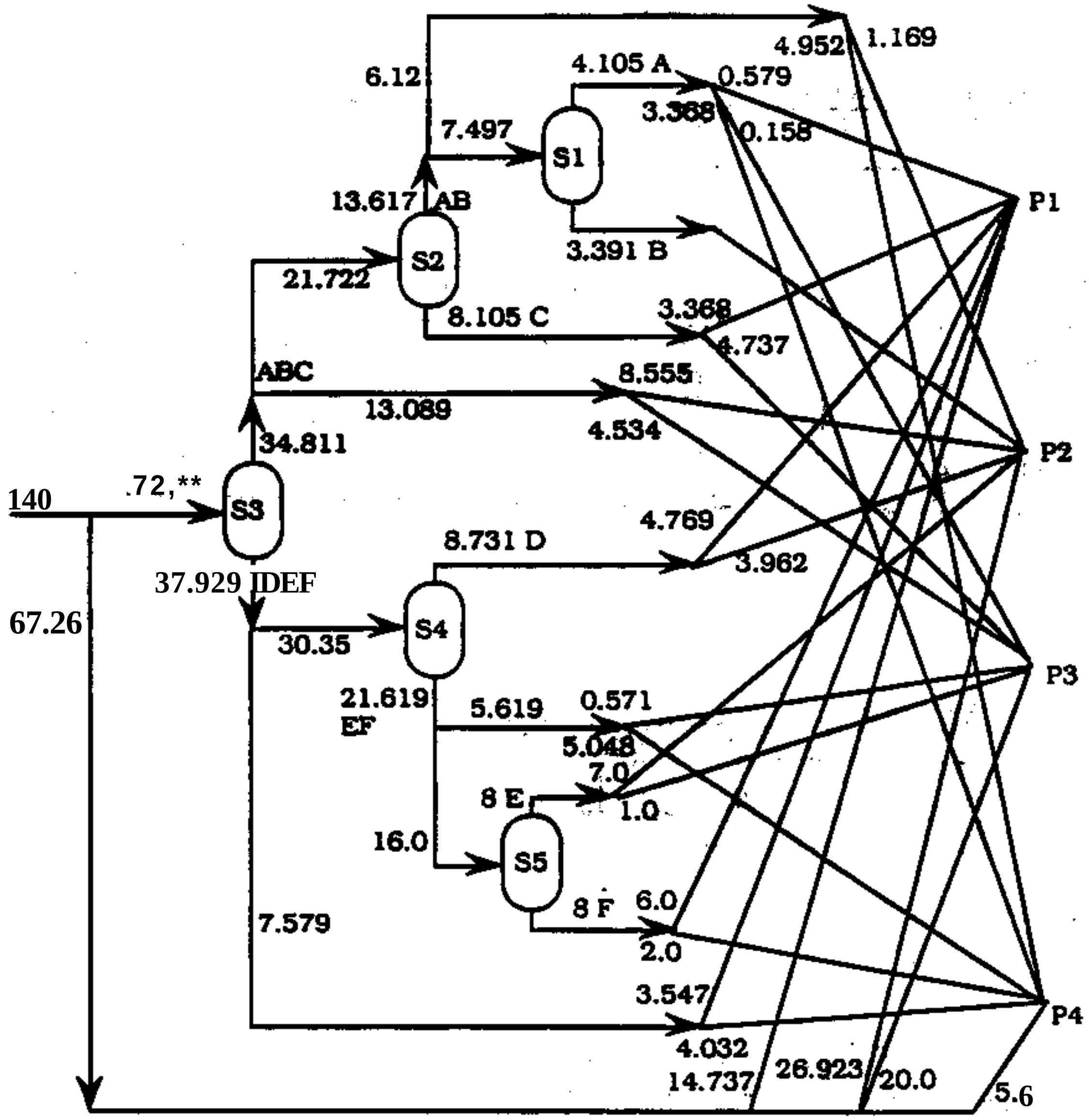

Figure 23. Solution of example 12.

Table 1. Computational results.

	Comp.	WocL	Var.	Lower bound	Initial gap	Global solution	Nodes	LP time	NLP time
Example 1	—	...	29	-500	20'	-400	3	0.05	0.1
Example 2	—	...	35	-513.22	0.3	-511.87	3	0.26	0.3
Example 3			113	138.18	0.4	138.7	1	0.34	0.4
Example 4	3	2	65	1.8639	0.0	1.8639	1	0.13	-.
Example 5	3	2	65	16	0.0	16	1	0.13	..
Example 6	4	2	107	54.25	2.3	55.5	3	0.97	0.4
Example 7	4	2	107	32.7	0.0	32.7	1	0.17	..
Example 8	4	3	125	26.76	0.1	26.79	1	0.23	0.3
Example 9	5	4	281	85.16	0.5	85.65	1	3.08	2.8
Example 10	5	2	225	156.56	12.4	159.48	5	2.59	2.3
Example 11	6	2	350	173	3.5	179.11	5	9.98	8.8
Example 12	6	4	430	362	14.8	388	33	19.8	13.2

Table 2. Data for example 6.

Component	A	B	C	D	Total
Product 1	5	10	4	10	29
Product 2	10	10	6	5	31
Feed	15	20	10	15	60

Table 3. Data for example 7.

Component	A	B	C	D	Total
Product 1	7.5	10		10	31.5
Product 2	7.5	10	6	5	28.5
Feed	15	20	10	15	60

Table 4. Data for example 8.

Component	A	B	C	D	Total
Product 1	2	3	1	3	9
Product 2	1	4	1	5	11
Product 3	3	1	3	1	8
Feed	6	8	5	9	28

Table 5. Data for example 9.

Component	A	B	C	D	E	Total
Product 1	7	8	3	9	8	35
Product 2	10	3	5	5	4	27
Product 3	5	5	6	7	3	26
Product 4	10	0	6	4	9	29
Feed	32	16	20	25	24	117

Table 6, Data for example 10,

Component	A	B	C	D	E	Total
Product 1	2	2.4	16	8	1	29.4
Product 2	8	5.6	4	8	9	34.6
Feed	10	8	20	16	10	64

Table 7. Data for example 11.

Component	A	B	C	D	E	F	Total
Product 1	3	2	16	8	4	10	4$
Product 2	8	10	8	8	6	5	4$
Feed	11	12	24	16	10	15	88

Table 8. Data for example 12.

Component	A	B	C	D	E	F	Total
Product 1	3	2	6	8	4	10	33
Product 2	8	10	8	8	6	5	45
Product 3	5	4	10	3	11	4	37
Product 4	7	3	1	2	5	7	25
Feed	23	19	25	21	26	26	140

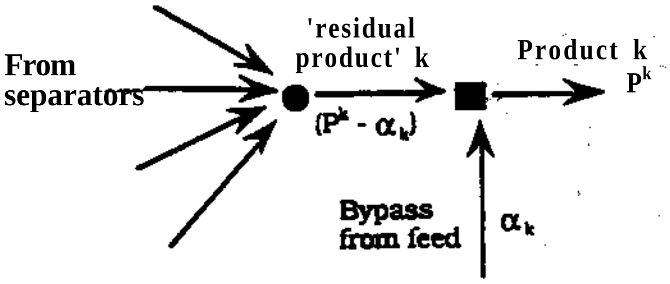

Figure 10. Definition of'residual product'.

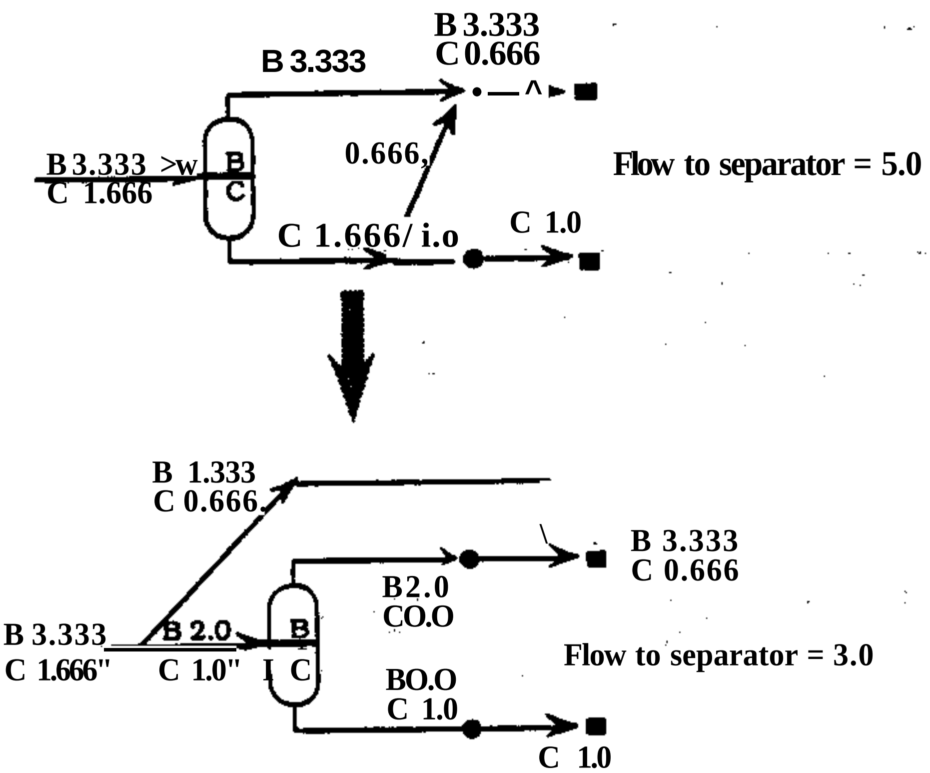

Figure 11. Example of solution without and with a zero component flow in 'residual product".

Global Optimization of Process Networks with Multicomponent Flows

I. Quesada, I.E. Grossman

EDRC 06-164-94

Global Optimization of Process Networks with Multicomponent Flows

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August 1993

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Abstract

This paper deals with the global optimization of networks consisting of splitters, mixers and linear process units and which involve multicomponent streams. Examples include pooling and blending systems and sharp separation networks. A reformulation-linearization technique is first applied to concentration and flow based models in order to obtain a relaxed LP formulation that provides a valid lower bound to the global optimum. This formulation is then used within a spatial branch and bound search. The application of this method is considered in detail for sharp separation systems with single feed and mixed products. Numerical results are presented on twelve test problems to show that only few nodes are commonly required in the branch and bound search.

Introduction

A common source of nonconvexities in the synthesis and design of processes, as well as in flowsheet optimization, are the material flow equations for multicomponent streams. These nonconvex equations involve bilinear terms and they arise in the mass balance equations when the compositions are unknown. There are different equivalent formulations for this type of networks. One alternative is to formulate the mass balance equations in terms of component concentrations* In this form bilinear terms are present in the equations for the mixer units and the different process units (e.g. sharp separators). A second alternative is to express the mass balances in terms of flows of individual components. This option has the advantage that it involves a smaller number of nonlinear equations. However, the modelling of the splitter units involves bilinear terms that arise due to the condition that the proportions of flows between components have to be the same for the different streams.

The difficulty with the nonconvexities noted above is that they may give rise to optimization problems involving several local optima and numerical singularities that may produce failure in the NLP algorithms. Recently there have been important efforts in the area of global optimization. Examples of algorithms are the ones proposed by McConnick (1976), Floudas and Viswewaran (1990) and Sherali and Alameddine (1992) which can be used to solve bilinear programming problems like the ones that arise in networks with multicomponent streams. For a recent review in the area of bilinear programming see Al-Khayyal (1992).

As for previous work in the design and synthesis of multicomponent process networks Mahalec and Motard (1977) and Nath (1977) developed evolutionary techniques that are based on heuristics to generate a network configuration. Floudas (1987) addressed the synthesis of separation networks with mixed products in which only sharp separators are considered. A superstructure of the process network was proposed and modelled in terms of concentrations. The resulting model is nonconvex and solved with a standard NLP algorithm with no guarantee of global optimality. Floudas and Aggaiwal (1990) solved small pooling and blending problems and sharp separation networks problems using a strategy based on Senders decomposition. In this approach only convex subproblems are solved but there is no guarantee of obtaining the global optimum. Kocis and Grossmann (1989) modelled process networks with multicomponent streams in terms of the individual component flows. They included a set of bounding constraints with binary variables to approximate the nonconvexities that are present in splitters with multiple outputs. Wehe and Westerberg (1987) studied the problem of sharp separation networks with mixed products. They proposed a search procedure that involves the

enumeration of the different separation sequences. The nonconvex equations are dropped and constraints that are valid for each particular sequence with a set of bounds over the key components are included to obtain tighter LP relaxations for each configuration. However, the number of sequences to be examined grows rapidly and there is no guarantee of global optimality*

In some particular cases the nonconvexities in the mass balances can be avoided through the introduction of binary variables. One of these cases is when single choice splitters are present in the flowsheet (Kocis and Grossmann. 1989). Here, it is possible to have a mixed integer ?fryfl»r formulation for the mass balance equations in terms of the individual component flows. Another restricted case for which some nonconvexities can be reformulated is when mining within the network is only allowed for streams of the same concentration. In this form, larger network superestructures must be proposed and the concentrations of the streams are known beforehand. Integer variables are introduced to model the existence of the different streams (e.g. the mixed integer linear formulation for sharp separation networks by Floudas and Anastasiadis, 1988).

The objective of this paper is to present an efficient global optimization method that exploits the particular structure that is present in process networks with multicomponent streams (e.g. pooling and blending systems, sharp separation networks). First a relation is established between formulations based on concentrations and individual flows. This is done following the Reformulation-Linearization technique proposed by Sherali and Alameddine (1992). As will be shown, a linear relaxation Is obtained that is in the space of the concentrations and individual flows which can be used in a branch and bound search is to find the global optimum. Application to the optimal design of sharp separation systems with single feed and mixed products is considered in detail. Different preprocessing techniques that allow tightening of the relaxation problem are presented. The performance of the algorithm is reported on a total of twelve problems.

Modeling with concentrations and individual flows

Consider a process network that consists of splitters, mixers and pfrbcess units tliat are interconnected with multicomponent streams (see Fig. 1). The process iinits that arc considered in this paper are units in which the output flows of the components can be expressed as a linear relation of the inlet flows (e.g. sharp separators, reactor with^known conversion). It is possible to formulate the mathematical model of the process network in terms of the concentrations of the streams, Xj^k. Another possibility is to model the network

using flows of individual components. The former has the advantages that it provides a convenient framework for the evaluation of thermodynamic properties, and in many cases bounds can be expressed in a more natural form. A major disadvantage is that many nonconvex terms (bilinear) are involved in the mass balances for the components. Hie individual component flows formulation is often chosen since it gives rise to a larger number of linear equations and the only nonconvexities are involved in the modelling of the splitters. In these untis it is necessary to enforce that the components maintain the same concentration in each of the streams leaving the splitter. These constraints can be expressed as relations between the different components (Wehe and Westerberg. 1987). One deficiency of this representation is that since many flows can take values of zero, singularities may arise with which conventional nonlinear programming methods may have difficulties to converge. Another alternative is to introduce additional variables that represent split fractions (Kocis and Grossmann. 1989). This involves a larger number of constraints but tends to yield a formulation that numerically is better behaved.

Following are the equations that apply to the mixers, splitters and units using the two alternative representations:

<u>Mixer</u>

A mixer k consists of a set of inlet streams, M_k, and an outlet stream k (see Fig.2).

a) Concentrations

The total mass balance for a mixer k is given by:

F*= 2 F* (1)

where F* Is the total flow in stream i. The mass balance for each component j is given by the nonlinear equations,

$F^{**} \wedge \sum_{i \in M_k} Pxj^1$ forallj (2)

where x_j^1 is the concentration of component j in the stream i

b) Individual flows

Here it is only necessary to write a mass balance for each component j, given by the linear equations:

$\wedge \wedge 2_{M_k} f_j^!$ forallj (3)

where fj^1 is the flow of component j in stream i.

<u>Splitter</u>

A splitter k has an inlet stream k and a set of outlet streams S* (see Fig. 3).

cQ Concentrations

The equations for a splitter in terms of the concentrations are given: by the following linear equations

$$F^{i} = F^{k} \quad (4)$$
$i \in S_k$

$$x_j^{i} = x_j^{k} \qquad \text{for alii 6 } S_k ai{*}dj \quad (5)$$

$$\sum_j x_j^{k} = 1 \quad (6)$$

'b) IndividuuaLJlows

The mass balance for each component J is given by

$$\sum_{i \in Sfc} f_j^{i}\ sff \qquad \text{forallj} \quad (7)$$

Here, it is also necessary to enforce the condition that the streams leaving the splitter have the same proportions in flow for each component. These relations between components, which are nonlinear, can be expressed in terms of the inlet stream k and a given component J*

$$fj{*}\ tf = fj^{k}\ fjV \qquad \text{for alii 6 } I_k andj{*}j' \quad (8)$$

A different approach consists of introducing as additional variables the split ratios ^, that represent the part of the inlet flow that goes to the outlet stream i. The nonlinear equations are given by

$$f_j^{i} = \xi^{i} f_j^{k} \qquad \text{(braliie } I_k andj \quad (9)$$

with $0 \le \xi^{i} \le 1$.

Process units

In this paper it will be assumed that the outlet streams, i € O_k, in the process units can be expressed as linear relations of the inlet streams, i e I_k (see Fig. 4). This is for instance the case of sharp split separators, separations in which the recovery level is known, or reactors that have a fixed conversion.

a) Concentrations

The overall mass balance for process unit k is given by,

$$IF{*}= \ £F \quad (10)$$
if Ik *«°k

The mass balance for each component] is given by the nonlinear equations:

$$£\ \ P{\wedge}F{*}'{\wedge}'sPx' \qquad \text{forallie } O_k andj \quad (11)$$
»*«lk

where p_{jr}^{lk} is a constant for process unit k that gives the distribution of component j in the stream i e O_k coming from streams i[1] € I_k. For a separator unit it is required tliat $\sum_{i \in O_k}$ f^* = 1 and £ Pji^{tk} s i. A sharp split separator is one for which $|I_k| = 1$ and $|O_k| = 2$ (top and bottom streams) and for all the components the constant p_{jt}^{ilc} are either 0 or 1.
$V \in I_k$

ffi Uvdiiikixialjlows

Only the **mas** balance for each component is necessary and it is given by:

$$\beta_{jr}^{ik} f_j^{r} \qquad \text{forallie } O_k \text{andj} \qquad (12)$$

A model in terms of individual flows MF consists of the linear equations (3), (7) and (12) plus the nonlinear equations (8) or (9). The model in terms of the concentrations. MX includes the linear equations (1). (4), (5). (6) and (10) plus the nonlinear equations (2) and (11).

Refor"? TI totton and Linearization

In order to avoid the direct use of the nonconvex models MX and MF, there is a relation that can be established between them using the reformulation and linearization technique for bilinear programming models proposed by Shcrali and Alameddine (1992). This technique can be applied to the model MX. First, consider the bounds over the variables present in the bilinear terms (total flow, F^1 and concentrations x_j^1)

$$F^{iL} \leq F^i \leq F^{iu} \qquad (13)$$

$$x_j^{iL} \leq x_j^i \leq x_j^{iu} \qquad (14)$$

Using the bounds in (13). (14) the following constraints can be generated for the bilinear terms in (2) and (UK

$$F^i x_j^i \geq F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

$$F^i x_j^i \geq F^{iu} x_j^i + x_j^{iu} F^i - F^{iu} x_j^{iu} \qquad (16)$$

$$F^1 X_j^4 \text{ £ } F^{*u} x_j^4 + x_j^{L} F^* - F^{*u} X_j^{IL} \qquad (17)$$

$$F^1 x_j^f \leq F^{iL} x_j^i + X_j^{4u} F^1 - F^{1L} X_j^{*u} \qquad (18)$$

In fact. McCormick (1976) has shown that the constraints in (15M 18) correspond to the convex and concave envelopes of the bilinear terms over the given bounds. The formulation is linearized by the definition of the following variables:

$$\$*¥**\{\bullet \qquad (19)$$

Tfte resultihg model which involves equations (1). (3), (4). (5), (6). (10). (12) and the constraints in (15)-{18) is a linear relaxation of the original nonconvex concentration model. MX. in which the nonlinear equations (2) and (11) have been replaced by the linear equations (3) and (12) from the individual flow model. MF. It is possible to generate additional linear constraints that are redundant to the original nonlinear model, MX, but that can be nonredundant in the linear relaxation of the model (Sherali and Alameddine, 1992: Sherali et

al.. 1992). In particular, consider equation (7) that is the linear component mass balance for the sputters in model MF. This linear equation is not present in the linear relaxation of the concentration model, MX. Take equation (4) and multiply by the valid bound constraint xf £ 0 to get

$$\sum_{i \in S_k} F x_j \wedge F * \wedge \qquad (20)$$

Using equation (5) yields.

$$\sum_{i \in S_k} P x_j \wedge F \wedge \qquad (21)$$

that can be linearized to,

$$\sum_{i \in S_k} f_j^i \; {}_{n} f_{i} k, \qquad (22)$$

yielding equation (7). Hence, the linear equation for the splitter is valid and it is Included. Hie nonlinear equations (8) or (9) can also be generated in a similar fashion but their linearizations are in general redundant (see Appendix A). They are only useful when the formulation df the problem provides non-trivial bounds over certain components in the outlet streams of a splitter, or when there exist some restrictions over the split ratios for the outlet streams.

Also, the constraints that relate the total flow and the individual flows of a stream can be generated for the splitters. Taking equation (6) and multiply by F^1 yields.

$$F^t \sum_j x_j^k = P \qquad (23)$$

Using the constraints $xf = x_j^k$ in equation (23) and linearizing with $f_j^1 = F^1 xf$ yields.

$$F\text{-}I t f_j \qquad (24)$$

Based on the above it is possible to obtain a reformulated model MR that involves concentrations, total flows and component flows, and which bounds the solution of the original problem. The following equations are given for model MR:

a) Objective function. 0. which is expressed in terms of individual or total flows, ..

b) Mixer equations, which are expressed ta terms of the total and individual component flows.

$$F^k = \sum_{i \in M_k} F^* \qquad (1)$$

$$f_j^k = \sum_{t \in M_k} f_j^\wedge \quad \text{for all } j \qquad (3)$$

c) Splitter equations, that are expressed in terms df the individual component flows and the concentrations of the streams

$$F^{l} = F^{k} \qquad l \in S_k \tag{4}$$

$x_j^{l} = x_j^{k}$ **for all i€ $S_k$andJ** **(5)**

$$\sum_{j} x_j^{k} = 1 \tag{6}$$

$\sum_{l \in S_k} f_j^{l} = f_j^{k}$ **forallJ** **(7)**

d) Process units equations, that are given in terms of the total and individual component flows

$\sum_{l \in Ifc} F^{l} = \sum_{l \in Ofc} F^{l}$ **(10)**

$f_j^{l} = \sum_{r \in I_k} \beta_{jr}^{lk} f_j^{r}$ **for all ie $O_k$andj** **(12)**

e) Relation between the total flow and the individual component flows

$F^{l} = \sum_{J} f_j^{l}$ **for all streams** **(24)**

f) Linear estimators, relate the individual component flows with the total flow and concentrations.

$f_j^{l} \geq F^{lL} x_j^{l} + x_j^{lL} F^{l} - F^{lL} x_j^{lL}$ **(15)**

fji S F ^ + x ^ P - F 1 " ^ " **foralli€S_k** **(16)**

fji ≤ jiu ^i ^ x^tL pi. pu x^iL **i s k € splitters** **t!7)**

$f_j^{l} \leq F^{lL} x_j^{l} + x_j^{lu} F^{l} - F^{lL} x_j^{lu}$ **foralljeJ** **(18)**

fj Bounds on flows and concentrations

$$F^{lL} \leq F^{l} \leq F^{lu} \tag{13}$$

$$x_j^{lL} \leq x_j^{l} \leq x_j^{lu} \tag{14}$$

In previous approaches (Wehe and Westerberg, 1987; Kocis and Grossmann, 1989) looser approximations of the nonlinear terms were used. In both cases, the nonconvex problem (MF) was relaxed to a linear model by dropping the nonlinear equations (8) or (9). Equations that approximate the difference relation between the components were considered (Kocis and Grossmann, 1989). They were based on the difference that exists at the inlet of the splitter between the flowrate of the components and required the introduction of binary variables.

Outline of global optimization method

Model MR can be applied to predict lower bounds to the global optimum in the optimization of pooling and blending problems and in the synthesis of separation systems. The reason is that model MR provides a valid relaxation of the original feasible region since the nonlinear equations (2) and (11) in model MX are not considered, and the valid linear equations (3), (7), (12) and (15)-(18) are included. The proposed global optimization algorithm relies on the solution of the relaxed problem MR within a spatial branch and bound enumeration. The outline of the algorithm is as follows (for a more detailed description of step 4 see Quesada and Grossmann (1993))

0. Preprocessing (optional)

Determine bounds on the variables involved in the nonconvex terms, that is total flows. F^*. and concentrations, *xf*. Apply any additional preprocessing specific to the structure of the problem in order to further bound or fix variables.

1. Lower Bound

Solve model MR over a given subregion (initial subregion is the complete feasible region) minimizing a convex objective function Φ. If Φ is linear the model is an LP.

2. *Upper Bound*

Any feasible solution to the nonlinear model provides an upper bound. Heuristic techniques can be employed to obtain good feasible solutions or the original problem. MF. can be solved using the solution of model MR as a good initial point. If the solution of problem MR is feasible it provides an upper bound.

3. *Convergence*

If the lower bound of a subregion is sufficiently close or above the upper bound discard that subregion. If no subregions are left the global solution corresponds to the best upper bound.

4. *Branch and Bound*

Partition the remaining subregions into a sell of disjoint subproblems. Repeat steps 1-3 for each of the new subregions.

Remarks

The preprocessing step plays an important role in the above algorithm. It is during this step that initial bounds for the variables involved in nonconvex terms are obtained. The quality of

these bounds affects the tigthness of the lower bound since they are part of the estimator equations (15)418). Additionally, these bounds affect the performance of the algorithm because they define the search space over which the branch and bound procedure may have to be conducted.

In some cases, as described later in this paper, it is possible to exploit the particular structure of the process network and generate bounds for the variables without having |o solve any subproblems. Furthermore, during this preprocessing step additional constraints can be generated for predicting a tighter lower bound of the global optimum can be obtained.

Some of the linear mass balances and the estimator equations are redundant in the nolinear formulations, MF and MX. These equations become nonredundant in the refonnulated model, MR, and for that reason it is important to write a complete formulation of the network. However, this model can present some redundancies that can be easily identified and eliminated to reduce the size of the model. This is the case for the concentration variables used in the splitters. Model MR uses different sets of concentrations variables for the inlet and outlet variables of a split unit. In practice, it is only necessary to define the concentration of the component in the splitter and use the same variables for all the splitter streams. Also, some redundancies can occur with the total flow variables. These ones are necessary for the streams in the splitters but they might be redundant and eliminated in the other untis if they do not appear in other part of the model or in the objective function.

If the solution of model MR is feasible for the original nonconvex problem then it corresponds to the global optimal solution. When the solution to the model MR is not feasible it is necessary to follow a branch and bound procedure to search for the global optimum. This procedure requires a valid upper bound on the global optimum. This can be generated through heuristics or by solving directly the nonconvex model. For this purpose, the process network model is formulated in terms of the individual component flows and the nonconvex equations for the splitters are included. Equation (9J was also used in this work to model the splitters due to it is better numerical behavior. The solution to the model MR was used for the good initial point. In many instances, it was not possible to solve these nonlinear problems with MINOS 5.2. The nonlinear models were solved using CONOPT in GAMS 2.25.

During the branch and bound procedure a tree search is generated. Of the set of open nodes, these are the nodes that have a lower bound that is e-smaller than the current upper bound, the node with the smaller upper bound is selected to branch on. The splitter units are the units that are approximated, and of these, the splitter that has the largest difference

between its approximated and actual individual component flows is selected. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. The branching is done in the selected splitter over the concentration of the component that has the largest difference.

First, the actual concentrations for the individual component flows in the LP solution O for the splitters are calculated by,

$$z_j^k = \frac{f_j^{k*}}{F^{k*}} \quad \text{for all the inlet streams to splitter} \tag{26}$$

the splitter unit m is then selected according to the equation.

$$m = \arg\max_{k \in split} \{ \sum_{i \in S_k} \sum_{j=1} | f_j^i - z_j^k F^i | \} \tag{27}$$

Equation (27) represents the total difference between the LP solution for the flows after the splitter and the actual value of these flows considering the concentrations before the splitter. Once the splitter has been selected, the component in that sputter that has the largest difference. J\ is selected by.

$$j^f = \text{argmax},(\sum_{i \in S_m} If/ -1£ FM] \tag{28»}$$

Hie following branching constraints are then used:

$$x_{j'}^m \leq z_{j'}^m \text{ and } Xj.^m \geq z,^m \tag{29}$$

To improve the upper bound it might be necessary to solve additional nonconvex problems. These can be solved after a given number of nodes using the solution of the node with the smallest upper bound as the initial point. In this work if there was no significant change in the lower bound df the new nodes with respect to the lower bound of the parent node (< 1%) a new nonconvex problem was solved.

Example 1

Consider the following pooling and blending problem by Harveley (1978). Two streams that have components A and B are mixed in a initial mixer a then go through a splitter to obtain two streams than can be mixed with an additional stream (see Fig. 5a). Two different products can be obatined and there are constraints on the concentration of component A in these products. The objective function consists of mimizing the cost that is given by the total flow of the streams times the cost coefficients. c_t, given in Fig. 5.

This problem has two local solutions. One has an objective function Φ = -100 and consists of *otify* producing product 2. The other local solution, that corresponds to the global optimum, has an objective of 4> = -400 and here only product 1 is produced.

Model MR is formulated for this problem and the initial lower bound is Φ_L = -500. The nonlinear model, MF, is solved using the solution of model MR as the initial point and an upper bound of • a -400 is obtained* Since there is a gap between the bounds of the global solution a partition is performed- There is only one splitter that needs to be approximated and since there are only two components it is irrelevant which one is selected since the composition bounds are related (eg. x_B^L s i - x^{*u}). The actual value of the composition of A in the solution of model MR is used as the branching point (x* = 0.0166) to generate two new subproblems. The first subproblem (x_A £ 0.0166) has a lower bound of Φ_L = -100 and the second subproblem (x* ≤ 0.0166) has a solution of fa = -400 (see Fig. 5b). Both of these bounds are greater or equal than the upper bound, therefore the global solution has been found (x_A = 0.01).

Example 2

Tbe next example is a separation problem taken from Kocis and Grossmann (1987). The original problem has binary variables in* the formulation and they have been fixed to 1 for this example (see Fig. 6).

The top stream of the flash unit has 85% of the inlet flow of A and the bottom stream has 80% of the inlet flow of component B. In the column. 97.5% of the inlet flow of A goes to the top whereas 95% of the inlet flow of B goes to the bottom stream. The total flow to the flash unit and the column have to be greater than 2.5 and smaller than 25, whereas the total flow of each of the two feed streams has to be less than 25. The objective function is given by,

$$\Phi \text{ ss } 52 + 10 F! + 8 F_2 + F_4 + 4 F_5 - 35 P,^A - 30 P_2^B \qquad (30)$$

The initial lower bound for this problem is Φ_L = -513.22 and it is infeasible for the original NLP model. A nonconvex problem is solved using CONOPT with the solution of model MR as the initial point obtaining an upper bound of Φ = -511.87 and the relative gap is only 0.3%. Again only one splitter is present in the network and a partition can be performed using the concentration of component A in this splitter. The lower bounds for the new two subproblems are 0L = -511.87 (x_A £ 0.5121) and 0_L = -511.80 ($x_A \geq$ 0.5121). Both solutions are greater or equal than the upper bound and the global solution has been obtained. In the global solution F_x ss 8 and F_2 = 25, and 11% of the inlet flow to the splitter is directed to the flash. 76% to the column and the rest bypassed to P_2.

Example 3

This example corresponds to a separation problem with three feeds and three product streams. The network configuration and product specifications are given in Fig. 7. The objective function is given by

$$\phi = 4\,S_1 + 1.5\,S_2 + 4\,S_3 \qquad (31)$$

The initial lower bound is ϕ_L = 138.18 and the nonconvex problem MP is solved obtaining a solution of ϕ « 138.7. The gap between these bounds is less than 0.4 %. The global optimum for this tolerance is shown In Fig. 8.

Sharp separation networks

In order to illustrate the application of the above algorithm to a specialized case where the structure can be further (exploited, the problem that will be considered is the synthesis of separation networks with single feed and mixed products that consist of sharp separators and bypasses. It is assumed that a single feed with N components must be separated into K specified multicomponent product streams. The components are ordered from the lightest to the heaviest.

A modification of the superstructure proposed by Floudas (1987) for this problem is used (see Fig. (9)). The superstructure consist of N-l separators. Separator 1 performs the task of removing component number 1 to number 1 at the top of the separator and components number i+1 to N at the bottom of the separator. The feed to the network is split into N-l streams, F_t. that go to the separators and K streams, a_k. that bypass the network to go to the products. Each stream F< is mixed before the separator i with streams that come from the top and bottom streams from the other separators to obtain the inlet stream to separator i, St.

The outlet streams of separator i are the top. T_{l9} and the bottom, B_t. These streams, T« and B{, are each split into streams, PT_t^k and PB<^{k} respectively, that go to the K products and into streams, KIV' and RB/\ that are redirected to the other separators. The top stream of separator 1, T_t, can be redirected only to the separators 1 to i-1 since it can only contain components number 1 to i. It would not be optimal to send part of this stream to any separator from i+1 to N since no separation would be achieved and a bypass of these separators would achieve the same separation with smaller (lows. *KTf* is the flow redirected from the top of separator i to separator i\ In the same fashion that with the top stream, the bottom of separator i, B_t, can be redirected only to separators i+1 to N-l since it can only

contain components that are separated by these sharp separators. $RB_i^{i'}$ is the redirected flow from the bottom stream of separator i to separator i'.

Model

It will be assumed that the objective function can be expressed as a linear function that depends on the total flow to each separator. The model expressed in terms of concentrations and total flows has the following form:

$$\min \Phi = \sum_{i=1}^{N-1} c_i S_i \qquad (32.1)$$

$$\text{s.t.} \quad Feed = \sum_{i=1}^{N-1} F_i + \sum_{k=1}^{K} \alpha_k \qquad (32.2)$$

$$f_{ij} = F_i z_j \quad \text{for all i and j} \qquad (32.3)$$

$$S_i = F_i + \sum_{i'=1}^{i-1} RB_{i'}^{i} + \sum_{i'=i+1}^{N-1} RT_{i'}^{i} \quad \text{for all i} \qquad (32.4)$$

$$S_i xs_{ij} = f_{ij} + \sum_{i'=1}^{i-1} RB_{i'}^{i} xb_{i'j} + \sum_{i'=i+1}^{N-1} RT_{i'}^{i} xt_{i'j} \quad \text{for all i and j} \qquad (32.5)$$

$$S_i = T_i + B_i \quad \text{for all i} \qquad (32.6)$$

$$T_i xt_{ij} = S_i xs_{ij} \quad \text{for all i and ord(j)} \le i \qquad (32.7)$$

$$xt_{ij} = 0 \quad \text{for all i and ord(j)} > i \qquad (32.8)$$

$$B_i xb_{ij} = S_i xs_{ij} \quad \text{for all i and ord(j)} > i \qquad (32.9)$$

$$xb_{ij} = 0 \quad \text{for all i and ord(j)} \le i \qquad (32.10)$$

$$T_i = \sum_{i'=1}^{i-1} RT_i^{i'} + \sum_{k=1}^{K} PT_i^k \quad \text{for all i} \qquad (32.11)$$

$$B_i = \sum_{i'=i+1}^{N-1} RB_i^{i'} + \sum_{k=1}^{K} PB_i^k \quad \text{for all i} \qquad (32.12)$$

$$P^k = \sum_{i=1}^{N-1} PT_i^k + \sum_{i=1}^{N-1} PB_i^k + \alpha_k \quad \text{for all k} \qquad (32.13)$$

$$P_{kj} = \sum_{i=1}^{N-1} PT_i^k xt_{ij} + \sum_{i=1}^{N-1} PB_i^k xb_{ij} + \alpha_k z_j \quad \text{for all k and j} \qquad (32.14)$$

$$\sum_{ord(j)=1}^{i} xt_{ij} = 1 \quad \text{for all i} \qquad (32.15)$$

$$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1 \quad \text{for all i} \qquad (32.16)$$

$$\sum_{ord(j)=1}^{N} xs_{ij} = 1 \quad \text{for all i} \qquad (32.17)$$

$$S_i, T_i, B_i, F_i, RT_i^{i'}, RB_i^{i'}, PT_i^k, PB_i^k, \alpha_k, f_{ij}, xs_{ij}, xt_{ij}, xb_{ij} \ge 0$$

The parameters Feed, z_j, P^k and p_{kj} represent the total feed, composition of the feed, total flow of product k and component flow of component j in product k. respectively. The variables xsq, xt_{ij} and xb«j are the concentrations of component j in the inlet stream to separator i. top of separator i and bottom of separator i, respectively.

The objective function (32.1) is a linear function of the inlet flow to the separators. Equation (32.2) is the total mass balance in the inlet splitter and equation (32.3) is the component mass balance. Equations (32.4) and (32.5) are the total and component mass balances for the mixer i before the separator L The material balances for separator i are given by equations (32.6). that is the total mass balance for the separator, equations (32.7) and (32.8) that are the component balances for the top stream and indicate that nothing from components number 1+1 to N-l is in the top of the separator, and equations (32.9) and (32.10) that are the component mass balances for the bottom streams. Equations (32.11) and (32.12) are the overall mass balances for the splitters of the top and bottom streams after separator i. The equations that state that the concentrations of the outlet streams should be the same that the inlet stream in a splitter have been already substituted. Finally, equations (32.13) and (32.14) are the overall and component mass balances for the mixer for product k.

Model (32) corresponds to a formulation of the type of model MX where the distribution coefficients are known and restricted to 0 or 1. Some simplifications have been made to avoid including many irrelevant variables (e.g. not to define concentrations for the streams that go the top i to product k). Although, some of the linear constraints in this formulation are redundant, they can become nonredundant in the linear relaxation as will be shown in Example 4.

Equations (32.5), (32.7), (32*9) and (32.14) involve nonconvex terms. This model can be reformulated as in model MR by introducing individual component flows and the linear equations (15)-(18) and (7) according to the approach illustrated eaiiier to obtain a model in the form of model MR. The resulting reformulated model is as follows,

$$\min \ \Phi' = \sum_{i=1}^{N-1} c_i S_i \qquad (33.1)$$

$$\text{st} \ \ \text{Feed} = \sum_{i=1}^{N-1} F_i + \sum^{K} \alpha_k \qquad (33.2)$$

$$f_{ij} = F_i z_j \qquad \text{for all i and j} \qquad (33.3)$$

$$S_i = F_i + \sum_{i'=1}^{i-1} RB_{i'}{}^{i} + \sum_{i'=i+1}^{N-1} RT_{i'}{}^{i} \qquad \text{for all i} \qquad (33.4)$$

$$s_{ij} = f_{ij} + \sum_{i'=1}^{i-1} rb_{i'j}{}^{i} + \sum_{i'=i+1}^{N-1} rt_{i'j}{}^{i} \qquad \text{for all i and j} \qquad (33.5)$$

$$S_i = T_i + B_i \qquad \text{for all i} \qquad (33.6)$$

$$t_{ij} = s_{ij} \qquad \text{for all i and ord(j) £ i} \qquad (33.7)$$

$$t_{ij} = 0 \qquad \text{for all 1 and ord(j) > 1} \qquad (33.8)$$

$$b_{ij} = s_{ij} \qquad \text{for all i and ord(j) > i} \qquad (33.9)$$

$$t_{ij} = 0 \qquad \text{for all i and ord(j)} \leq \text{i} \qquad (33.10)$$

$$T_i = \sum_{i'=1}^{i-1} RT_i^{i'} + \sum_{k=1}^{K} PT_i^k \qquad \text{for all i} \qquad (33.11)$$

$$B_i = \sum_{i'=i+1}^{N-I} RB_i^{i'} + \chi PB_i^k \qquad \text{for all 1} \qquad (33.12)$$

$$P^k = {}^{N\text{-}I} PT_i^k + \sum_{i=1}^{N-1} PB_i^k + \alpha_k \qquad \text{for all k} \qquad (33.13)$$

$$p_{kj} = \sum_{i=1}^{rt\text{-}i} pt_{ij}^k + \sum_{i=1}^{N-1} pb_{ij}^k + \alpha_k z_j \qquad \text{for all k and J} \qquad (33.14)$$

$$t_{ij} = \sum_{i'=1}^{i-1} rt_{ij}^{i'} + \sum_{k=1}^{K} pt_{ij}^k \qquad \text{for alii and j} \qquad (33.15)$$

$$b_{ij} = \sum_{i'=i+1}^{N-I} rb_{ij}^{i'} + \sum_{k=1}^{K} pb_{ij}^k \qquad \text{for all i and j} \qquad (33.16)$$

$$\sum_{ord(j)=1}^{i} xt_{ij} = 1 \qquad \text{for all i} \qquad (33.17)$$

$$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1 \qquad \text{for all i} \qquad (33.18)$$

$$T_i = \sum_{ord(j)=1}^{i} t_{ij} \qquad \text{for all i} \qquad (33.19)$$

$$B_i = \sum_{ord(j)=i+1}^{N} b_{ij} \qquad \text{for alii} \qquad (33.20)$$

$$PT_i^k = \sum_{ord(j)=1}^{i} pt_{ij}^k \qquad \text{for all i and k} \qquad (33.21)$$

$$PB_i^k = \sum_{ord(j)=i+1}^{N} pb_{ij}^k \qquad \text{for all 1 and k} \qquad (33.22)$$

'' '' •-S/ (i) for all i and i' < i (33.23)

$$RB_i^{i'} = \sum_{ord(j)=i+1}^{N} rb_{ij}^{i'} \qquad \text{for all i and i'} > \text{i} \qquad (33.24)$$

Equations (15-18) forty. rt_{rj}^{1} and pt_{y}^{k} in terms of xtg and the total flow of its respective stream.

Equations (15-18) for by. rbq^{1} and pb_{tl}^{k} in terms of xb_{ij} and the total flow of its respective stream.

S,.T,, B,. F,. RT,'. RB,^{f}. PT,^{k}. PB,^{k}, Ok. f_{tt}. xs,j. *xU$_{t}$*. $xb_{(J}$ SO

Sg. t,j. b,j pti,^{k}. pb,,^{11}. rt,,$^{1'}$. rb,,$^{1'}$ ≥ 0

It is not necessary to Include equations (15)-(18) for the inlet component flows to the separator, Sq, since the variables xs,j only appear in these equations. Also, the component flows, s,j, only appear in mixers and sharp separators units which can be exactly represented in terms of the individual component flow equations (33.5). (33.7) and (33.9). Equations (33.15)-(33.16) that are the component mass balances for the splitters of top and bottom streams have been included accordingly to the reformulation previously presented. Equations (33.19M33.24) relate the total flow and the individual component flows for the splitter streams.

Preprocessing

Hie proposed superstructure (Fig. 9) allows to bypass certain amount of the feed to the product k, a*, without having to go through the separation network. The amount of the product k that is not bypassed has to be processed in the separation network and it will be denoted as the 'residual product*. Hence, the total 'residual product k' is given by (P^k - a_k) and has the component flows given by (p^ - o^z,)(see Fig. 10).

The global optimal solution of model (32) is a network in which all the 'residual products'.have at least one component with a zero flow. The reason that it is not optimal to separate a stream in the network and later to remix it. The same degree of separation can be achieved using a bypass that does not incur any cost in the objective function.

Consider the second separator in the solution obtained by Floudas (1987) to his second example (see Fig. 11). For this subnetwork of the complete structure the upper 'residual product* has components B and C present. The components are being separated and remixed again. The same outlet flows can be obtained with a smaller input flow to the separator as it is shown in Fig. 11. Note that both 'residual products' have components with zero flow.

It should be clear that if there was not a component with zero flow in the 'residual product[1], then there is part of this stream that could have been obtained by just bypassing the network. This in turn does not incur in any cost, whereas going through the network has a positive cost. The above condition gives a lower bound for the bypass to each product. This also corresponds to the largest amount that can be bypassed since all the flows in the 'residual flow* have to be positive. In this form the bypass can be precalculated without affecting the global optimality of the solution.

The bypass to product k is given by the maximum amount that can be sent to product k without having a negative flow; that is.

a„ = min, [^4 (34)

where z_j is the feed composition and p_{kj} is the flow of component j in product k. The component flows for 'residual product' k. γ_{kj} are given by.

$$\gamma_{kj} = p_{kj} - \alpha_k z_j \qquad (35)$$

Key component bounds

Wehe and Westerberg (1987) proposed using lower bounds for the (low of the key components in separator L These bounds are based on the fact that separator i is the only unit that can perform the task of separating component number 1 from component nutaber 1+1. They are redundant for the nonlinear formulation (32) but they are relevant for the linear relaxation in (33). To calculate them, it is necessary to determine in each product what is the difference between the two key components of separator i with respect to the concentrations in the feed. The lower bounds in separator i' for the flow of the key components in the top (component $j1$) and bottom (component $j2$) streams are given by:

$$t_{ij1} \geq \sum_{k=1}^{K} \{ \gamma_{kj1} - z_{j1} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (36)$$

$$b_{ij2} \geq \sum_{k=1}^{K} \{ \gamma_{kj2} - z_{j2} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (37)$$

where γ_{kj} is given by (35). It is important to include both bounds in the relax model (33) since there is no guarantee that the inlet flow to separator i has the same proportion between the key components as the feed. It is not known in which part of the sequence separator i will be placed, and it can be after a splitter that is not being approximated correctly.

The bounds in (36) and (37) can be extended to separation of components that are not adjacent in the feed. Consider component number i and component number i+3. There are three separators that can perform this task/separators i. i+1 and i+2. Cuts of the following form can be obtained.

$$t_{i\,i} + t_{i+1\,i} + t_{i+2\,i} \geq \sum_{k=1}^{K} \{ \gamma_{ki} - z_i \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (38)$$

$$b_{i\,i+3} + b_{i+1\,i+3} + b_{i+2\,i+3} \geq \sum_{k=1}^{K} \{ \gamma_{k\,i+3} - z_{i+3} \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (39)$$

Equations as the ones in (38) and (39) can be redundant compared to equations (36) and (37) and it is possible to detect this before solving the problem.

Relative flowrate constraints

These constraints are used when the relation between the flowrates of two components is known. In particular, consider component A in the la:st column of the network (see Fig. 12). None of the redirected streams contains component A. Therefore, the relative flowrate of component A with respect to the other components in the top stream has to be smaller than in the feed. This relation should remain valid after the top stream is split to the products and redirected flows.

In the separator previous to the last one, N-2, all the streams do not have component A except the one coming from the top of the last column. For this one it is already known that the relative flow of component A with respect to the other components is smaller than in the feed. This type of analysis can be done for component A and component N in all the columns yielding th$ following linear constraints for the splitters.

for all i and k

$$z_A pt_{ij}^k - z_j pt_{iA}^k \ge 0 \qquad \text{for } j \ne A \text{ and ord(j)} \le i \qquad (40)$$

$$z_N pb_{ij}^k - z_j pb_{iN}^k \ge 0 \qquad \text{for ord(j)} \ne N \text{ and ord(j)} > i \qquad (41)$$

for all i and i' > i

$$z_A rt_{ij}^{i'} - z_j rt_{iA}^{i'} \ge 0 \qquad \text{for } j \ne A \text{ and ord(j)} \le i \qquad (42)$$

for all i and i' < i

$$z_N rb_{ij}^{i'} - z_j rb_{iN}^{i'} \ge 0 \qquad \text{for ord(j)} \ne N \text{ and ord(j)} > i \qquad (43)$$

Bounds on concentrations and total Jlows

The approximations (15-18) require bounds for the total flows and component concentrations In the splitters. The lower bound for the total flow of the top and bottom streams is given by the lower bound of the key components obtained in equations (36) and (37). For the outlet streams of the splittters, that are the redirected streams and the streams that go to the products, the lower bound is zero. The upper bound for the total flow of the top and bottom streams is given by the feed to the network of the components that can be present in each stream .that is.

$$T_i^U = \sum_{ord(j)=1}^{i} Feed \cdot \sum_k \alpha_{ik} z_j, \qquad \text{for all i} \qquad (44)$$

$$B_i^U = \sum_{ord(j)=i+1}^{N} Feed \cdot \sum_k \alpha_{ik} z_j \qquad \text{for all i} \qquad (45)$$

The upper bound for the streams after the splitter are given by.

$RT_i^{i' u} = T_i^u$ for all i and $i' < i$ (46)

$RB_i^{i' u} = B_i^u$ for alii aridi'> i (47)

$PT_i^k = \sum_{and(j)=1}^{i} Y_{kj}$ for all i and k (48)

$PB_i^k =$ Jb? for all i and k (49)

The lower bounds for the concentrations are zero except for fee key components in the separator for which the lower bounds are given by the lower bound of its flow divided by the upper bound of the total flow of that stream. The upper bounds in the concentrations are given by one minus the lower bounds of the other components.

The solution of the linear programming model (33) provides a lower bound to the global optimum since this model is a valid relaxation of the nonconvex model (32). This lower bound is obtained by solving the LP model for the 'residual products' in (35) with the addition of the valid constraints (36)-(43).

The upper bounds are generated solving model (32) in terms of the individual flows for the 'residual products'. When additional nonconvex problems are solved to improve the current upper bound it can happen that very similar initial point are generated. In this case, a new nonconvex NLP is solved in which bounds over the total inlet flows to the separators (S_i) are included. For this purpose the values of these variables in the LP solution (S_i^*) arc used such that the current incumbent solution is no longer feasible.

Example4

Consider the 3 component example proposed by Floudas and Aggarwal (1990). An equimolar feed has to be separated into two products as show in Fig. 13. The objective function is given by

$$\phi = 0.2395 + 0.00432\ S_1 + 0.7584 + 0.01517\ S_2 \qquad (50)$$

The bypass to products 1 and 2 can be calculated according to equation (34) and the 'residual product' component flows are obtained through equation (35) (see Fig. 14). The total bypass to product 1 is $\alpha_1 = 90$ and the bypass to product 2 is $\alpha_2 = 150$ and the feed has a concentration of $z_A = 1/3$, $z_B = 1/3$ and $z_C = 1/3$. In this form the 'residual product' 1 is $Y_{1A} = 0$, $Y_{1B} = 20$ and $Y_{1C} = 0$ and the 'residual product' 2 is $Y_{2A} = 20$, $Y_{2B} = 0$ and $Y_{2C} = 20$. Additionally, lower bounds on the flow of the key components in both separators are obtained

using equations (35M36). The key components in separator 1 are component A in the top and its flow has to be at least 20 and component B in the bottom has to have at least a flow of 20. In the top stream of the second separator at least 20 units of component B have to be separated from 20 units of component C in the bottom stream. It is important to note that after preprocessing the network several suboptimal solutions have been cut off. One of these suboptimal solutions for this particular data is a parallel configuration of both separators (there are situations in which a parallel configuration can correspond to the global solution as will be shown in example 5). In this example the direct or indirect sequence have a lower objective function. Both of these configurations are local solutions with an objective function value of 4 = 1.8639 for the direct sequence and $ = 2.081 for the indirect one. In some instances, MINOS 5.2 had problems converging even in this small example.

The LP (33) is formulated for this problem, giving a lower bound of ^ = 1.8639. The approximations are exact and therefore this solution is a feasible solution of model (32) proving that it corresponds to the global optimum. Hence, convergence is achieved in one iteration. The optimum solution corresponds to the direct sequence shown in Fig. 15. It should be noted that if the linear mass balances for the mixer for product 2 were not considered since they are redundant for the nonlinear formulation, a lower bound in the relaxed model of $\$_L$ = 1.12 is obtained. This shows that It is relevant to include all the mass balances in the linear model in order to tighten the lower bound.

ExompteS

In the approach proposed by Wehe and Westerberg (1987) for the case of 3 components only the direct and indirect sequences are considered and both options can be modelled as LP problems since no mixing is required for these separation networks. However, this example shows that parallel configurations can be also globally optimal and that they are not excluded by the method proposed in this paper. To be able to consider parallel configurations or any combination of parallel with direct or indirect sequences it is necessary to model a superstructure in which mixing is allowed (like in the structure used in Fig. 13). Here, nonconvexities arise in the mass balance equations after the separators.

Consider that an equimolar feed is to be separated into the two different products given in Fig. 16. The objective function is to minimize the sum of the total flows into the separators. The same procedure that in the previous example is followed and the bypass can be precalculated by equation (34). The solution to the model (32) yields Φ_L = 12 and since it is a feasible solution of model (32) it corresponds to the global optimum (see Fig. 16). Note that the solutions for the direct or indirect sequences have an objective function of $ = 16.

Branch and Bound

If there is a gap between the lower ai}d upper bound, a branch and bound search is performed. It is only necessary to do the search over the variables involved in the nonconvex terms. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. In this way. it is necessary to check the approximation for the concentrations in the splitters of the top and bottom strear^s of the separator. Equations (26)-(29) for the splitters of top and bottom streams are used to perfom the branch and bound search.

Results

Table 1 summarizes the results of the earlier examples 1 to 3 and of the sharp separation network examples 4 to 12. The number of variables is the total number of variables that are included in the reformulated and relaxed model (33) for that example. The lower bound is the initial bound that is obtained by solving model (33) over the entire feasible space. The inital gap represents the percentage difference between the initial lower and upper bounds. When there is a zero initial gap it means that the first relaxed solution is feasible in the original problem thereby corresponding to the global solution. The column for nodes gives the total number of subproblems that where solved before converging to the global solution. A relative tolerance of 0.01 was used, except for example 2 were exact convergence was obtained after branching and example 12 for which a tolerance of 0.02 was used. It is important to note that the initial lower bound is tight and that it corresponds to a good estimation of the global solution. The largest differences are for example 1 with a 25% of difference and for example 12 with a 7% difference. The LP time refers to the time used to solve each relaxed model and the NLP time is the time used for solving a nonconvex model. It is possible to do updates using the previous LP solution and in this form have a more efficient implementation. The times are in seconds and the problems were solved on an IBM RS600/530 using GAMS 2.25 (Brooke et al. (1988)). MINOS 5.2 was used to solve the LP problems and CONOPT for the nonconvex NLP problems. A brief description of the example problems 6 to 12 is given below. It includes the specific data for the problem, the objective function and the topology of the network that is the global solution.

Example 6

This example corresponds to example 2 from Floudas (1987). In this case a linear objective function with the same cost coefficients is used and it is given by,

$$(M2.5S!+3.0S_2+1.5S_3 \qquad (51)$$

The data for the composition of the products is given in Table 2.

The initial lower bound is ϕ_L = 54.25 and an upper bound of $ = 55.5 is obtained by solving the nonconvex problem. A partition of the feasible region is performed using the composition of component D in the bottom stream of separator 1. The first subproblem (x £ 0.166) yields a lower bound of ^ = 55.45 and the second subproblem (x < 0.166) has a solution of ϕ_L= 55.8. The latter is greater than the upper bound and the former is less than 1% of the global solution (see Fig. 17).

Exampie7

This example is taken from Floudas (1987). The data for this problem is given in Table 3 and the linear objective function is given by:

$$\phi = 2.5\, S_1 + 3.0 S2 + 1.2 S_3 \qquad (52)$$

The initial lower bound is ϕ_L = 32.7 and it provides a feasible solution to the nonconvex problem. In this form the global solution (see Fig. 18Ms obtained in one iteration. It is interesting to see that this solution also provides a better objective function for the concave objective function used by Floudas (1987) (4 = 10.65 versus 0 = 13.68 which is 28% higher)

Elxample8

This four component problem is taken from Wehe and Westerberg (1987). The data for the products is given in Table 4 and the objective function has the following form:

$$\phi \, s\, 5.0 + 0.5\, Si + 4.0 + 0.3\, S2 + 6.0 + 0.7\, S_3 \qquad (53)$$

The first relaxed subproblem has a solution of ϕ_L =26,76 and it is infeasible for the nonconvex problem. A nonconvex problem is solved using CONOPT with the LP solution as the initial point. An upper bound of 0 « 26.79 is obtained corresponding to the global solution (see Fig. 19) within a 0.1%.

Example 9

This example corresponds to example 1 from Wehe and Westerberg (1987). Table 5 provides the data for the product flows and the objective function is given by:

$$\phi = 5.0 + 0.5\, Si + 9.0 + 1.0\, S2 + 3.0 + 0.4\, S_3 + 6.0 + 0.6\, S_4 \qquad (54)$$

A initial lower bound of ϕ_L = 85.16 is obtained and the upper bound Is 0 » 85-65. The difference is 0.5% and the global solution (see Fig. 20) is obtained in one iteration.

Example 10

This problem is taken from Flotidas (1987) and the data is given in Table 6. The objective function is given by,

$$\Phi = 1.2\,S_1 + 3.0\,S_2 + 2.5\,S_3 + 1.5\,S_4 \qquad (55)$$

The initial lower bound is Φ_L = 156.56 and the upper bound is Φ = 179.08. After 5 nodes the global solution of Φ = 159.48 (see Fig. 21) is obtained.

Example 11

Hie data for this 6 component problem are given in Table 7 and the objective function has the following form:

$$\Phi = 1.5\,S_1 + 3.0\,S_2 + 2.0\,S_3 + 1.0\,S_4 + 4.0\,S_5 \qquad (56)$$

The initial lower bound is Φ_L = 173 and the upper bound is Φ = 179.11. After five nodes the global solution is obtained (see Fig. 22).

Example 12

This is a 6 component 4 products problem and the data are given in Table 8. The objective function is:

$$\Phi = 5.0\,S_1 + 3.0\,S_2 + 2.0\,S_3 + 2.5\,S_4 + 4.0\,S_5 \qquad (57)$$

The initial lower bound is Φ_L = 362 and the initial upper bound is Φ = 415.6. The global solution of Φ = 388 (with a 2% optimality gap) is obtained after 33 nodes (see Fig. 23).

Conclusions

A general procedure for the global optimization of process networks with multicomponent streams has been proposed. The basic idea relies on a relaxed LP model that is obtained through reformulation-linearization techniques that establish a clear relation between the component flow and the composition models for mass balances. The reformulated model combines both of these providing tighter lower bounds than other relaxations proposed in the previous work. The relaxed linear model has been embedded in a branch and bound procedure to obtain the global optimal solution.

As has also been shown, the algorithm can be further specialized to take advantage of the particular structure of sharp separation networks with single feed and mixed products. Here, it is possible to preprocess the problem to reduce the space over which the search is

conducted. The bounds that are necessary for the estimator functions in the relaxed model can be obtained without having to solve any subproblems. Different types of linear approximations that are nonredundant to the relaxed model are included to obtain a tighter lower bound.

Twelve examples for both general process networks and for sharp separation networks have been presented to illustrate the performance of the algorithm. As has been shown, only a small number of nodes are commonly needed in the branch and bound search to identify the global or e-global solution. Moreover, in many cases the initial lower bound is either the exact solution or a very good approximation to the global solution.

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Appendix A. Reformulation-Linearization to obtain the nonlinear constraints in **model MF**

The nonlinear equations in **model** MF, that can be expressed either as (8) or (9), can also be generated from model MX. For this purpose take the concentration model MX and consider equation (5),

... $x_j^k = x_j^l$ (A.1)

multiply by the valid bound constraint Xj-k £ 0

xfxfmxfzp **(A.2)**

Use equation (5) for **component** j\

x,kVil*fk** **1A-3'**

Multiply by **the valid bound** constraints $F^k \geq 0$ and $F^1 \geq 0$,

F^JP^-F^F*^ tA.41

that it is linearized to yield.

fffJJsfj'ff (A.5)

which is precisely equation (8) for the splitters in the individual flow model MF.

Consider again equation (5).

$x_j^k = x_j^1$ (A.6)

multiply by the valid bound constraints $F^k \geq 0$ and $F^* \geq 0$,

$F^k x_j^k F^l = F^l x_j^l F^k$ (A.7)

that can be linearized to yield,

ffF^f/F* (A.8)

Define the split fraction ? to be,

?=|* (A.9)

Equation (A.8) can then be expressed as

f_j^l = ? f_j^k (A.10)

which corresponds to equation (9).

Hence, the nonlinear equations (8) and (9) are redundant to model MX. Their linear approximations in general are also redundant in the linear reformulated model MR Consider equation (A.1O), similarly to (15) one of its linear approximations is given by.

$$f_j^l \geq \xi^{lL} f_j^k + \xi^l f_j^{kL} - \xi^{lL} f_j^{kL} \quad \text{(A.11)}$$

If there are no particular restrictions in the splitters, then the bounds for the split fraction variable are $0 \leq \xi \leq 1$ and using them in (A. 11) yields.

$$f_j^1 \geq f_j^{kL} \xi^1 \quad \text{(A.12)}$$

The bound for the individual component flow is given by $f_j^{kL} = x_j^L F^{kL}$: also $x_j^k = x_j^4$ and $\xi^f = \frac{F^1}{F^t}$, which leads to.

$$f_j^1 \geq x_j^{L} F^1 \frac{F^{kL}}{F^k} \quad \text{(A.13)}$$

The estimator (15) for the same conditions ($F^{*L} = 0$) is given by

$$f_j^1 \geq x_j^{L} F^1 \quad \text{(A.14)}$$

Since the factor $\frac{F^{kL}}{F^k}$ is always less or equal than 1, equation (A. 13) is redundant. A similar analisis can be performed for the other estimators. Only when more specific bounds over the split fractions or the individual component flows are known, will these additional estimators be non redundant.

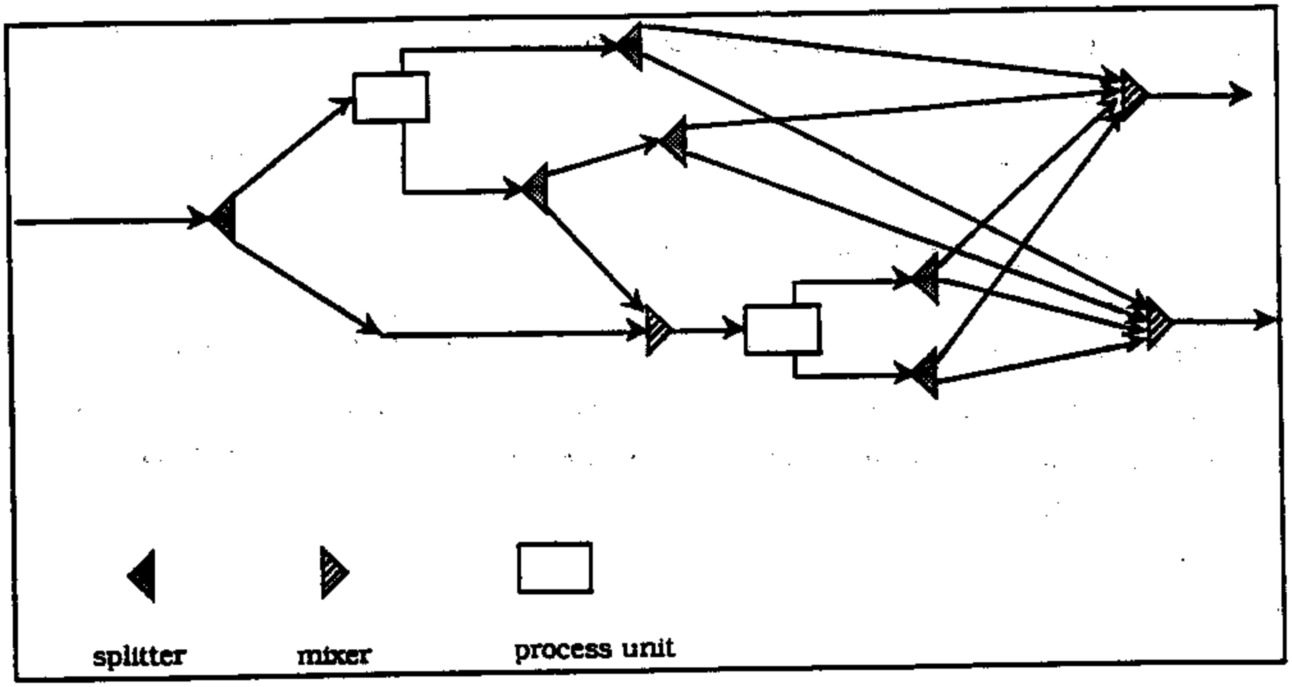


Figure 1. Process network with units, splitters and mixers.

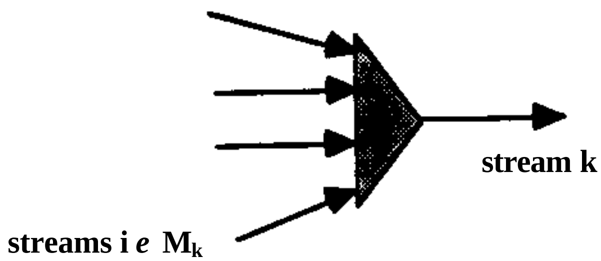


Figure 2. Mixer module.

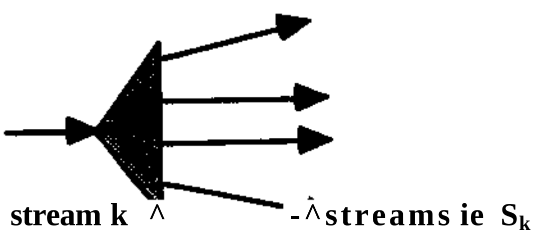


Figure 3. Splitter module.

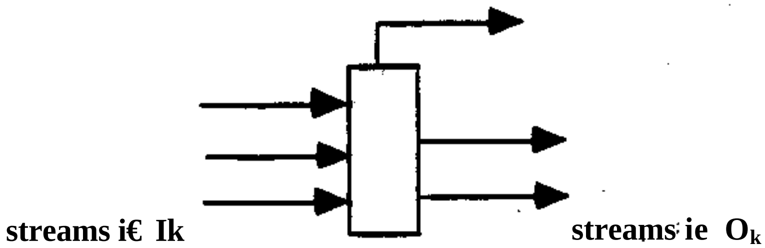
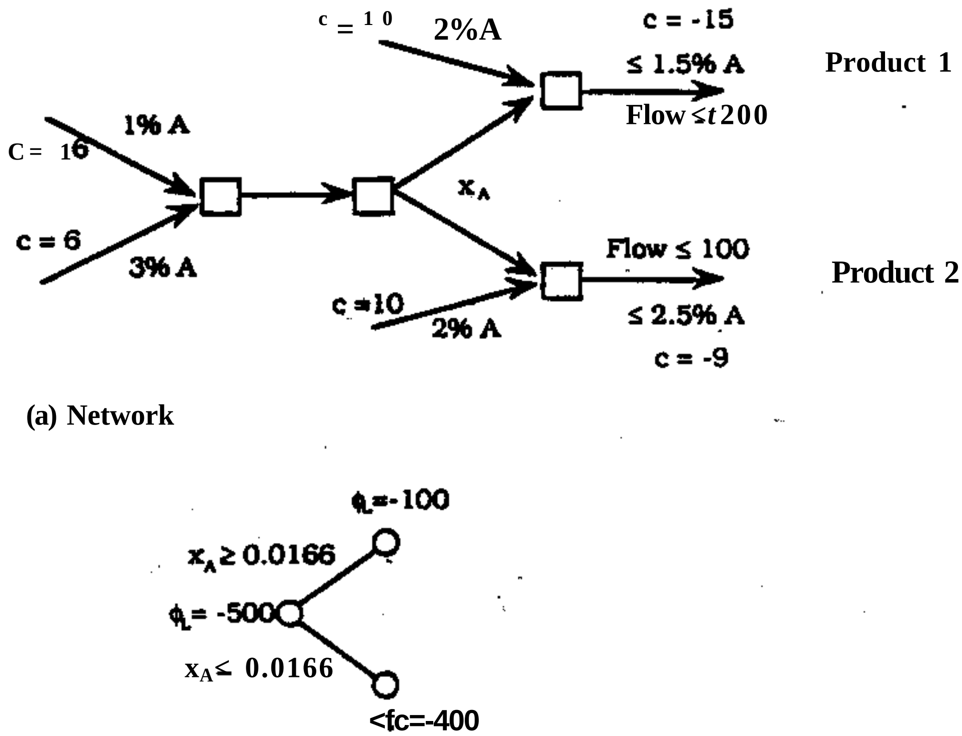


Figure 4. Process unit module.



(a) Network

(b) Tree search

Figure 5. Network and branch and bound search for example 1

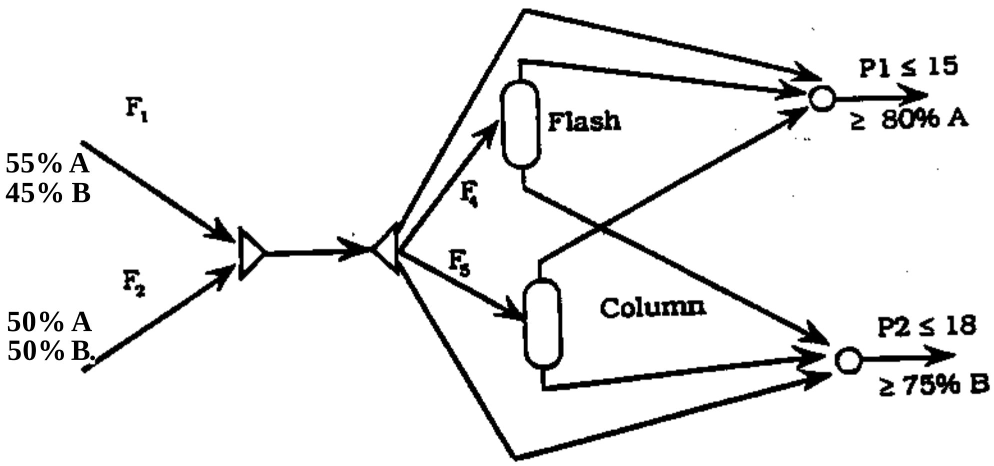


Figure 6. Network for example 2.

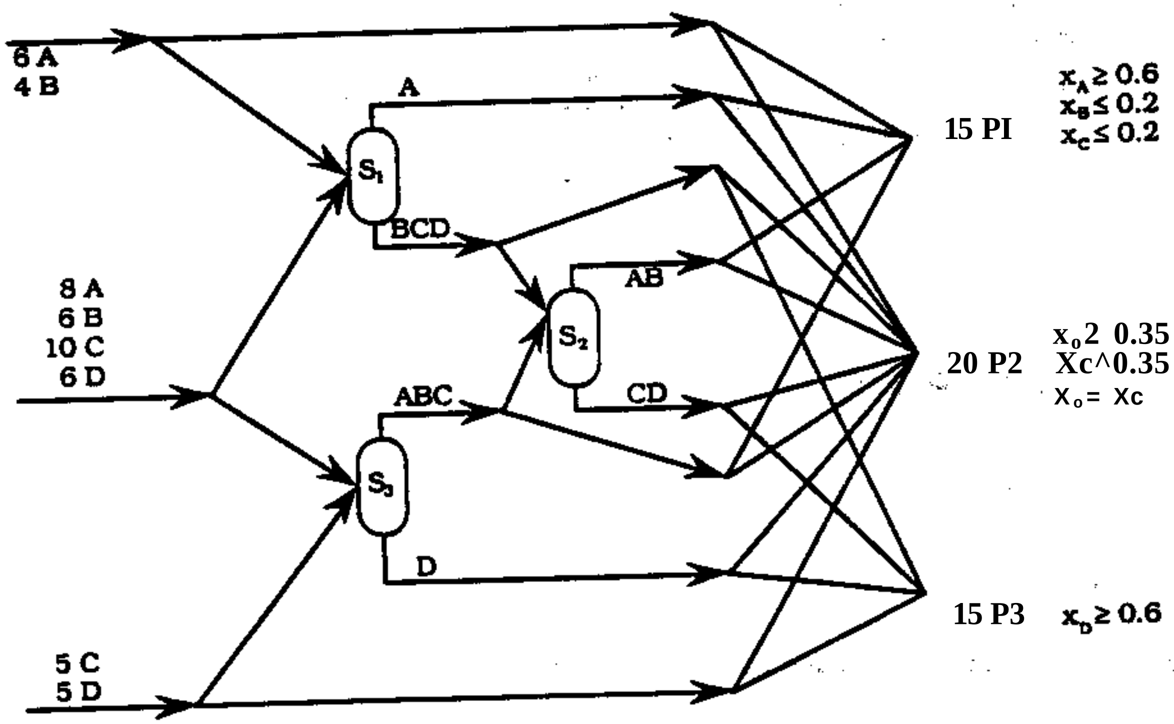


Figure 7. Network for example 3.

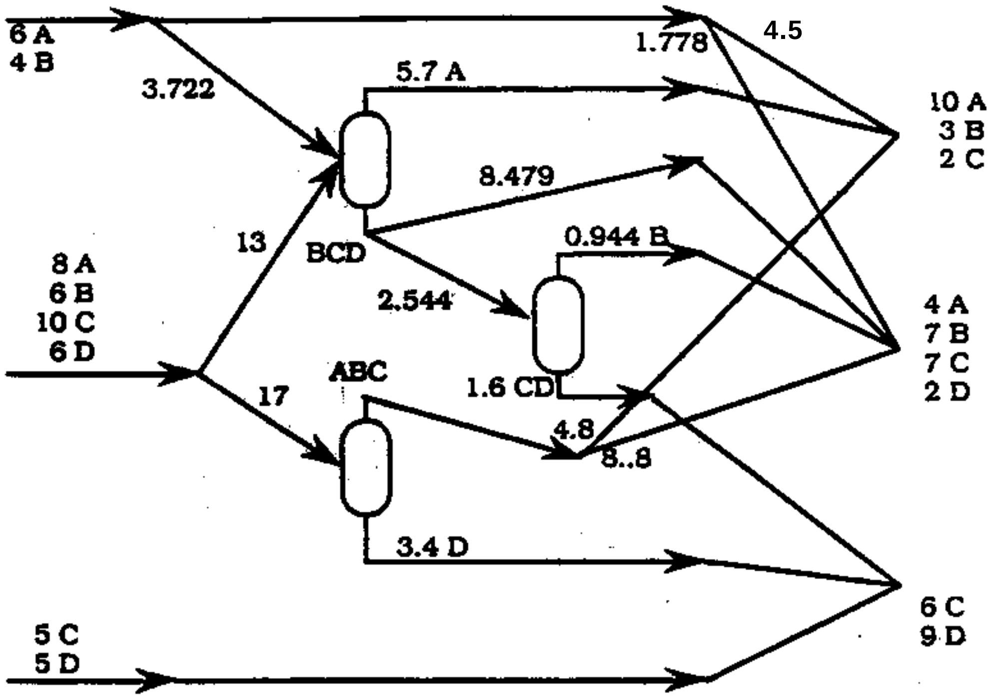


Figure 8. Optimal network for example 3.

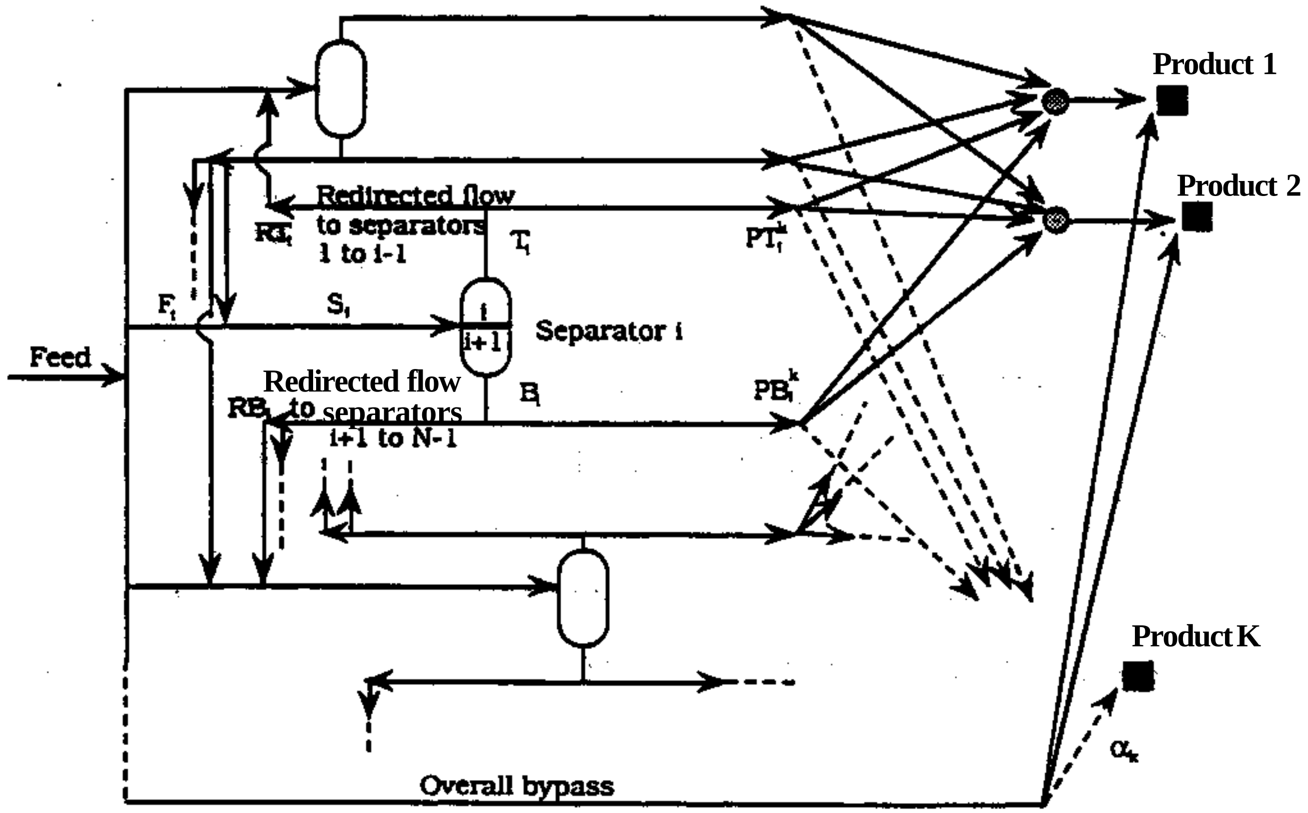


Figure 9. Superstructure for separation with sharp splits and mixed products.

Global Optimization of Process Networks with Muiticomponent Flows

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Global Optimization of Process Networks with Multicomponent Flows

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Abstract

This paper deals with the global optimization of networks consisting of splitters, mixers and linear process units and which involve multicomponent streams. Examples include pooling and blending systems and sharp separation networks. A reformulation-linearization technique is first applied to concentration and flow based models in order to obtain a relaxed LP formulation that provides a valid lower bound to the global optimum. This formulation is then used within a spatial branch and bound search. The application of this method is considered in detail for sharp separation systems with single feed and mixed products. Numerical results are presented on twdve test problems to show that only few nodes are commonly required in the branch and bound search.

Introduction

A common source of noneonvexities in the synthesis and design of processes, as well as in flowsheet optimization, are the material flow equations for multiccmponent streams. These nonconvex equations involve bilinear terms and they arise in the mass balance equations when the compositions are unknown. There are different equivalent formulations for this type of networks. One alternative is to formulate the mass balance equations in terms of component concentrations. In this form bilinear terms are present in the equation* for the mixer units and the different process units (e.g. sharp separators). A second alternative is to express the mass balances in terms of flows of individual components. This option has the advantage that it involves a ^*naUmr* number of nonlinear equations. However, the modelling of the splitter units involves bilinear terms that arise due to the condition that the proportions of flows between components have to be the same for the different streams.

The difficulty with the nonconvexities noted above is that they may give rise to optimization problems Involving several local optima and numerical singularities that may produce failure in the NLP algorithms. Recently there have been important efforts in the area of global optimization. Examples of algorithms are the ones proposed by McCormick (1976). Floudas and Viswewaran (1990) and Sherali and Alameddine (1992) which can be used to solve bilinear programming problems like the ones that arise in networks with multicomponent streams. For a recent review in the area of bilinear programming see Al-Khayyal (1992).

As for previous work in the design and synthesis of multicomponent process networks Mahalec and Motard (1977) and Nath (1977) developed evolutionary techniques that are based on heuristics to generate a network configuration. Floudas (1987) addressed the synthesis of separation networks with mixed products in which only sharp separators are considered. A superstructure of the process network was proposed and modelled in terms of concentrations. The resulting model is nonconvex and solved with a standard NLP algorithm with no guarantee of global optimality. Floudas and Aggarwal (1990) solved small pooling and blending problems and sharp separation networks problems using a strategy based on Benders decomposition. In this approach only convex subproblems are solved but there is no guarantee of obtaining the global optimum. Kocis and Grossmann (1989) modelled process networks with multicomponent streams in terms of the individual component flows. They included a set of bounding constraints with binaxy variables to approximate the nonconvexities that are present in splitters with multiple outputs. Wehe and Westerberg (1987) studied the problem of sharp separation networks with mixed products. They proposed a search procedure that involves the

enumeration of the different separation sequences. The nonconvex equations are dropped and constraints that are valid for each particular sequence with a set of bounds over the key components are included to obtain tighter UP relaxations for each configuration. However, the number of sequences to be examined grows rapidly and there is no guarantee of global optimality.

In some particular cases the nonconvexities in the mass balances can be avoided through the introduction of binary variables. One of these cases is when single choice splitters are present in the flowsheet (Kocis and Grossmann, 1989). Here, it is possible to have a mixed integer linear formulation for the mass balance equations in terms of the individual component flows. Another restricted case for which some nonconvexities can be reformulated is when mixing within the network is only allowed for streams of the same concentration. In this form, larger network superestructures must be proposed and the concentrations of the streams are known beforehand. Integer variables are introduced to model the existence of the different streams (e.g. the mixed integer linear formulation for sharp separation networks by Floudas and Anastasiadis, 1988).

The objective of this paper is to present an efficient global optimization method that exploits the particular structure that is present in process networks with multicomponent streams (e.g. pooling and blending systems, sharp separation networks). First a relation is established between formulations based on concentrations and individual flows. This is done following the Reformulation-Linearization technique proposed by Sherali and Alameddine (1992). As will be shown, a linear relaxation Is obtained that is in the space of the concentrations and individual flows which can be used in a branch and bound search is to find the global optimum. Application to the optimal design of sharp separation systems with single feed and mixed products is considered In detail. Different preprocessing techniques that allow tightening of the relaxation problem are presented. The performance of the algorithm is reported on a total of twelve problems.

Modeling with concentrations and individual flows

Consider a process network that consists of splitters, mixers and process units that are interconnected with multicomponent streams (see Fig. 1). The process iinits that are considered in this paper are units in which the output flows of the components can be expressed as a linear relation of the inlet flows (e.g. sharp separators, reactor with^known conversion). It is possible to formulate the mathematical model of the process network in terms of the concentrations of the streams. Xj^k. Another possibility is to model the network

using flows of individual components. The former has the advantages that it provides a convenient framework for the evaluation of therxnodynamic properties, and in many cases bounds can be expressed in a more natural form. A major disadvantage is that many nonconvex terms (bilinear) are involved in the mass balances for the components. The individual component flows formulation is often chosen since it gives rise to a larger number of linear equations and the only nonconvexities are involved in the modelling of the splitters. In these untis it is necessary to enforce that the components maintain the same concentration in each of the streams leaving the splitter. These constraints can be expressed as relations between the different components (Wehe and Westerberg, 1987). One deficiency of this representation is that since many flows can take values of zero, singularities may arise with which conventional nonlinear programming methods may have difficulties to converge. Another alternative is to introduce additional variables that represent split fractions (Kocis and Grossmann. 1989). This involves a larger number of constraints but tends to yield a formulation that numerically is better behaved.

Following axe the equations that apply to the mixers, splitters and units using the two alternative representations:

<u>Mixer</u>

A mixer k consists of a set of Inlet streams, M_k, and an outlet stream k (see Fig.2).

a) Concentrations

The total mass balance for a mixer k is given by:

$$F^k = \sum_{i \in M_k} F^i \qquad (1)$$

where F^i is the total flow in stream i. The mass balance for each component j is given by the nonlinear equations.

$$F^k x_j^k = \sum_{i \in M_k} F^i x_j^i \qquad \text{for all j} \qquad (2)$$

where x_j^i is the concentration of component j in the stream i

b) IndividualJlows

Here it is only necessary to write a mass balance for each component j, given by the linear equations:

$$f_j^k = \sum_{i \in M_k} f_j^i \qquad \text{for all j} \qquad (3)$$

where f_j^i is the flow of component j In stream i.

<u>Splitter</u>

A splitter k has an inlet stream k and a set of outlet streams S_k (see Fig. 3).

ql[Concentrations

The equations for a splitter in terms of the concentrations are given by the following linear equations

$$\sum_{1 \in S_k} P = P^* \qquad (4)$$

$$x_j^* = x_j^k \qquad \text{for all } i \in S_k \text{ and } j \qquad (5)$$

$$\sum_j x = 1 \qquad (6)$$

b) Individual flows

The mass balance for each component j is given by

$$\sum_{i \in S_k} f_j^i = f_j^k \qquad \text{for all } j \qquad (7)$$

Here, it is also necessary to enforce the condition that the streams leaving the splitter have the same proportions in flow for each component. These relations between components, which are nonlinear, can be expressed in terms of the inlet stream k and a given component j'

$$f_{j'}^k f_j^i = f_j^k f_{j'}^i \qquad \text{for all } i \in I_k \text{ and } j \neq j' \qquad (8)$$

A different approach consists of introducing as additional variables the split ratios ξ^i that represent the part of the inlet flow that goes to the outlet stream i. The nonlinear equations are given by

$$f_j^i = \xi^i f_j^k \qquad \text{for all } i \in I_k \text{ and } j \qquad (9)$$

with $0 \le \xi^i \le 1$.

Process units

In this paper it will be assumed that the outlet streams, $i \in O_k$, in the process units can be expressed as linear relations of the inlet streams, $i \in I_k$ (see Fig. 4). This is for instance the case of sharp split separators, separations in which the recovery level is known, or reactors that have a fixed conversion.

a) Concentrations

The overall mass balance for process unit k is given by,

$$\sum_{i \in I_k} P = \sum_{i \in O_k} P \qquad (10)$$

The mass balance for each component j is given by the nonlinear equations;

$$\sum_{i' \in I_k} \beta_{jr}^{ik} F^{i'} x_j^{i'} = P x_j^i \qquad \text{for all } i \in O_k \text{ and } j \qquad (11)$$

where P_{jr}^{ik} is a constant for process unit k that gives the distribution of component j in the stream $i \in O_k$ coming from streams $i' \in I_k$. For a separator unit it is required that $\sum_{i \in O_k} \beta^{ik} = 1$ and $\sum_{i' \in I_k} P_{jr}^{ik} = 1$. A sharp split separator is one for which $|I_k| = 1$ and $|O_k| = 2$ (top and bottom streams) and for all the components the constant p_{jr}^{ik} are either 0 or 1.

b) Individual flows

Only the mas balance for each component is necessary and it is given by:

$$f_j^i = \sum_{l \in I_k} \beta_j^{lk} f_j^{l} \qquad \text{for all } i \in O_k \text{ and } j \qquad (12)$$

A model in terms of individual flows MF consists of the linear equations (3), (7) and (12) plus the nonlinear equations (8) or (9). The model in terms of the concentrations, MX, includes the linear equations (1), (4), (5), (6) and (10) plus the nonlinear equations (2) and (11).

Reformulation and Linearization

In order to avoid the direct use of the nonconvex models MX and MF, there is a relation that can be established between them using the reformulation and linearization technique for bilinear programming models proposed by Sherali and Alameddine (1992). This technique can be applied to the model MX. First, consider the bounds over the variables present in the bilinear terms (total flow, F^i and concentrations x_j^i)

$$F^{iL} \le F^i \le F^{iU} \qquad (13)$$

$$x_j^{iL} \le x_j^i \le x_j^{iU} \qquad (14)$$

Using the bounds in (13), (14) the following constraints can be generated for the bilinear terms in (2) and (11),

$$F^i x_j^i \ge F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

$$F^i x_j^i \ge F^{iU} x_j^i + x_j^{iU} F^i - F^{iU} x_j^{iU} \qquad (16)$$

$$F^i x_j^i \le F^{iU} x_j^i + x_j^{iL} F^i - F^{iU} x_j^{iL} \qquad (17)$$

$$F^i x_j^i \le F^{iL} x_j^i + x_j^{iU} F^i - F^{iL} x_j^{iU} \qquad (18)$$

In fact, McConnick (1976) has shown that the constraints in (15)-(18) correspond to the convex and concave envelopes of the bilinear terms over the given bounds. The formulation is linearized by the definition of the following variables:

$$f_j^i = F^i x_j^i \qquad (19)$$

Tile resulting model which involves equations UK (3). (4). (5), (6). (10). (12) and the constraints in (15M18) is a linear relaxation of the original nonconvex concentration model, MX, in which the nonlinear equations (2) and (11) have been replaced by the linear equations (3) and (12) from the individual flow model. MF. It is possible to generate additional linear constraints that are redundant to the original nonlinear model, MX, but that can be nonredundant in the linear relaxation of the model (Sherali and Alameddine, 1992: Sherali et

al*. 1992). In particular, consider equation (7) that is the linear component mass balance for the splitters in model MF. "This linear equation is not present in the linear relaxation of the concentration model. MX. Take equation (4) and multiply by the valid bound constraint $x_j^k \geq 0$ to get

$$\sum_{i \in S_k} F^k x_j^k = F^k x_j^k \quad (20)$$

Using equation (5) yields.

$$\sum_{i \in S_k} F^i x_j^i = F^k x_j^k \quad (21)$$

that can be linearized to.

$$\sum_{i \in S_k} f_j^i = f_j^k. \quad (22)$$

yielding equation (7). Hence, the linear equation for the splitter is valid and it is included. The nonlinear equations (8) or (9) can also be generated in a similar fashion but their linearizations are in general redundant (see Appendix A). They are only useful when the formulation 6f the problem provides non-trivial bounds over certain components in the outlet streams of a splitter, or when there exist some restrictions over the split ratios for the outlet streams.

Also, the constraints that relate the total flow and the individual flows of a stream can be generated for the splitters. Taking equation (6) and multiply by F^1 yields.

$$F^i \sum_j x_j^k = F^i \quad (23)$$

Using the constraints $x_j^i = x_j^k$ in equation (23) and linearizing with $f_j^i = F^i x_j^i$ yields,

$$F^i = \sum_j f_j^i \quad (24)$$

Based on the above it is possible to obtain a reformulated model MR that involves concentrations, total flows and component flows, and which bounds the solution of the original problem. The following equations are given for model MR:

a) Objective function. ⊕ which is expressed in terms of individual or total flows.

b) Mixer equations, which are expressed in terms of the total and individual component flows.

$$F^k = \sum_{i \in M_k} F^i \quad (1)$$

$$f_j^k = \sum_{i \in M_k} f_j^i \qquad \text{for all } j \quad (3)$$

c) Splitter equations, that are expressed in terms of the individual component flows and the f^oncditi^ tf^nff of the streams

y p-pk (t€S_k) (4)

Xj' = Xj^k for alii *e* 3* and J (5)

XxJ<=i (6)

£ If «ff for ail j (7)

d) Process units equations, that are given in terms of the total and individual component flows

$$\sum_{i \in I_k} F^i = \sum_{i \in O_k} F^i \qquad (10)$$

fj«= T p/'f/ (r ∈ I_k) foraUie $O_k$andj (12)

e) Relation between the total flow and the individual component flows

$$F^i = \sum_j f_j^i \qquad \text{for all streams} \qquad (24)$$

f) Linear estimators, relate the individual component flows with the total flow and concentrations.

ri £> pi L jqi + x* F* pi . pi L jri L (15)

fji ^F^1tt^4^^!uF^l-F^lu^lu for alii e S_k (16)

f/ ≤F^ltt^l4-^1LF^i-F^!u^IL i s k € splitters flT)

fj^1 ^F^XJ' + X^F-F'-XJ'" foralljeJ (18)

f) Bounds on flows and concentrations

$$F^{iL} \le F^i \le F^{iu} \qquad (13)$$

$$x_j^{iL} \le x_j^i \le x_j^{iu} \qquad (14)$$

In previous approaches (Wehe and Westerberg, 1987; Kocis and Grossmann, 1989) looser approximations of the nonlinear terms were used. In both cases, the nonconvex problem (MF) was relaxed to a linear model by dropping the nonlinear equations (8) or (9). Equations that approximate the difference relation between the components were considered (Kocis and Grossmann, 1989). They were based on the difference that exists at the inlet of the splitter between the flowrate of the components and required the introduction of binaiy variables.

Outline of global optimization method

Model MR can be applied to predict lower bounds to the global optimum in the optimization of pooling and blending problems and in the synthesis of separation systems. The reason is that model MR provides a valid relaxation of the original feasible region since the nonlinear equations (2) and (11) in model MX are not considered, and the valid linear equations (3), (7), (12) and (15)-(18) are included. The proposed global optimization algorithm relies on the solution of the relaxed problem MR within a spatial branch and bound enumeration. The outline of the algorithm is as follows (for a more detailed description of step 4 see Quesada and Grossmann (1993))

0. Preprocessing (optional)

Determine bounds on the variables involved in the nonconvex terms, that is total flows, F^1, and concentrations, $x\{$. Apply any additional preprocessing specific to the structure of the problem in order to further bound or flx variables.

1. *Lower Bound*

Solve model MR over a given subregion (initial subregion is the complete feasible region) minimizing a convex objective function $. If ◊ is linear the model Is an LP.

2. Upper Bound

Any feasible solution to the nonlinear model provides an upper bound. Heuristic techniques can be employed to obtain good feasible solutions or the original problem, MF. can be solved using the solution of model MR as a good initial point. If the solution of problem MR is feasible it provides an upper bound.

3. *Convergence*

If the lower bound of a subregion is sufficiently close or above the upper bound discard that subregion. If no subregions are left the global solution corresponds to the best upper bound.

4. Branch and Bound

Partition the remaining subregions into a set of disjoint subproblems. Repeat steps 1-3 for each of the new subregions.

Remarks

The preprocessing step plays an important role in the above algorithm. It is during this step that initial bounds for the variables involved in nonconvex terms are obtained. The quality of

these bounds affects the tigthness of the lower bound since they are part of the estimator equations (15)418). Additionally, these bounds affect the performance of the algorithm because they define the search space over which the branch and bound procedure may have to be conducted.

In some cases, as described later in this papier, it is possible to exploit the particular structure of the process network and generate bounds for the variables without having to solve any subproblems. Furthermore, during this preprocessing step additional constraints can be generated for predicting a tighter lower bound of the global optimum can be obtained.

Some of the linear mass balances and the estimator equations are redundant in the nolinear formulations, MF and MX. These equations become nonredundant in the reformulated model, MR. and for that reason it is important to write a complete formulation of the network. However, this model can present some redundancies that can be easily identified and eliminated to reduce the size of the model. This is the case for the concentration variables used in the splitters. Model MR uses different sets of concentrations variables for the inlet and outlet variables of a split unit. In practice, it is only necessary to define the concentration of the component in the splitter and use the same variables for all the splitter streams. Also, some redundancies can occur with the total flow variables. These ones are necessary for the streams in the splitters but they might be redundant and eliminated in the other untis if they do not appear in other part of the model or in the objective function.

If the solution of model MR is feasible for the original nonconvex problem then it corresponds to the global optimal solution. When the solution to the model MR Is not feasible it is necessary to follow a blanch and bound procedure to search for the global optimum. This procedure requires a valid upper bound on the global optimum. This can be generated through heuristics or by solving directly the nonconvex model. For this purpose, the process network model is formulated in terms of the individual component flows and the nonconvex equations for the splitters are included. Equation (9) was also used in this work to model the splitters due to it is better numerical behavior. The solution to the model MR was used for the good initial point. In many instances, it was not possible to solve these nonlinear problems with MINOS 5.2. The nonlinear models were solved using CONOPT in GAMS 2.25.

During the branch and bound procedure a tree search is generated. Of the set of open nodes, these are the nodes that have a lower bound that is e-smaller than the current upper bound, the node with the smaller upper bound is selected to branch on. The splitter units are the units that are approximated, and of these, the splitter that has the largest difference

between its approximated and actual individual component flows is selected. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. The branching is done in the selected splitter over the concentration of the component that has the largest difference.

First, the actual concentrations for the individual component flaws in the LP solution (*) for the splitters are calculated by,

$z_j^k = \frac{f_j^k}{F^k}$ for all the inlet streams to splitter (26)

The splitter unit m is then selected according to the equation.

m = arg max$_k$ c mitt ($\sum_{S_k} \sum_{j=1}^{N}$ | ty-z^FH) , (27)

Equation (27) represents the total difference between the LP solution for the flows after the splitter and the actual value of these flows considering the concentrations before the splitter. Once the splitter has been selected, the component in that splitter that has the largest difference. J\ is selected by,

J--aigma^(JL"I$_{S_m}$ f/ - z»^{e} F* 11 (28)

The following branching constraints are then used;

$x_j^m \le z_j^m$ and $x_j^m \ge z_j^m$ (29)

To improve the upper bound it might be necessary to solve additional nonconvex problems. These can be solved after a given number of nodes using the solution of the node with the smallest upper bound as the initial point. In this work if there was no significant change in the lower bound of the new nodes with respect to the lower bound of the parent node (< 1%) a new nonconvex problem was solved.

Example 1

Consider the following pooling and blending problem by Harveley (1978). Two streams that have components A and B are mixed in a initial mixer a then go through a splitter to obtain two streams than can be mixed with an additional stream (see Fig. 5a). Two different products can be obatined and there are constraints on the concentration of component A in these products. The objective function consists of niimizing the cost that is given by the total flow of the streams times the cost coefficients, c_t, given in Fig. 5.

This problem has two local solutions. One has an objective function 0 = -100 and consists of only producing product 2. The other local solution, that corresponds to the global optimum, has an objective of + « -400 and here only product 1 is produced.

Model MR is formulated for this problem and the initial lower bound is 0_L = -500. The nonlinear model. MF, is solved using the solution of model MR as the initial point and an upper bound of $ s -400 is obtained. Since there is a gap between the bounds of the global solution a partition is performed. There is only one splitter that needs to be approximated and since there axe only two components it is irrelevant which one is selected since the composition bounds are related (eg. $x_B^L = 1 \bullet x^{*u}$). The actual value of the composition of A in the solution of model MR is used as the branching point (x* = 0.0166) to generate two new subproblexns. The first subproblcm (x_A £ 0.0166) has a lower bound of 4_L = -100 and the second subproblem (x* £ 0.0166) has a solution of fa. = -400 (see Fig. Sb). Both of these bounds are greater or equal than the upper bound, therefore the global solution has been found (x_A = 0.01).

Example 2

The next example is a separation problem taken from Kocis and Grossmann (1987). The original problem has binary variables in the formulation and they have been fixed to 1 for this example (see Fig. 6).

*

The top stream of the flash unit has 85% of the inlet flow of A and the bottom stream has 80% of the inlet flow of component B. In the column. 97.5% of the inlet flow of A goes to the top whereas 95% of the inlet flow of B goes to the bottom stream. Hie total flow to the flash unit and the column have to be greater than 2.5 and smaller than 25, whereas the total flow of each of the two feed streams has to be less than 25. The objective function is given by,

$$\Phi = 52 + 10 F! + 8 F_2 + F_4 + 4 F_5 - 35 Pj^A - 30 P_2^B \quad (30)$$

The initial lower bound for this problem is 0_L = -513.22 and it is infeasible for the original NLP model. A nonconvex problem is solved using CONOPT with the solution of model MR as the initial point obtaining an upper bound of Φ = -511.87 and the relative gap is only 0.3%. Again only one splitter is present in the network and a partition can be performed using the concentration of component A in this splitter. The lower bounds for the new two subproblexns are *fa* = -511.87 (x_A £ 0.5121) and Φ_L = -511.80 ($x_A \geq$ 0.5121). Both solutions are greater or equal than the upper bound and the global solution has been obtained. In the global solution Fi = 8 and F_2 = 25, and 11% of the inlet flow lo the splitter is directed to the flash. 76% to the column and the rest bypassed to P_2.

Example 3

TIUs example corresponds to a separation problem with three feeds and three product streams. The network configuration and product specifications are given in Fig. 7. The objective function is given by

$$>\ll 4 S i + 1.5 S2 + 4 S3 \qquad (3D$$

The initial lower bound is 4_L = 138.18 and the nonconvex problem MP is solved obtaining a solution of $ = 138.7. The gap between these bounds is less than 0.4 %. The global optimum for this tolerance is shown in Fig. 8.

Sharp separation networks

In order to illustrate the application of the above algorithm to a specialized case where the structure can be further exploited, the problem that will be considered is the synthesis of separation networks with single feed and mixed products that consist of sharp separators and bypasses. It is assumed that a single feed with N components must be separated into K specified multicomponent product streams. The components are ordered from the lightest to the heaviest.

A modification of the superstructure proposed by Floudas (1987) for this problem is used (see Fig. (9)). The superestructure consist of N-l separators. Separator i performs the task of removing component number 1 to number! at the top of the separator and components number i+1 to N at the bottom of the separator. The feed to the network is split into N-l streams, F_t, that go to the separators and K streams, a_k. that bypass the network to go to the products. Each stream $F_{\{}$ is mixed before the separator i with streams that come from the top and bottom streams from the other separators to obtain the inlet stream to separator i, S^

The outlet streams of separator i are the top, $T_{(}$, and the bottom, B_t. These streams, T, and B_t. are each split into streams, PT_t^k and PB^ respectively, that go to the K products and into streams, KIV' and.RBf. that are redirected to the other separators. The top stream of separator i, T_{it} can be redirected only to the separators 1 to i-1 since it can only contain components number 1 to i. It would not be optimal to send part of this stream to any separator from i+1 to N since no separation would be achieved and a bypass of these separators would achieve the same separation with smaller flows. *KTf* is the flow redirected from the top of separator i to separator i\ In the same fashion that with the top stream, the bottom of separator i, B,, can be redirected only to separators i+1 to N-l since it can only

contain components that are separated by these sharp separators. RB/ is the redirected flow from the bottom stream of separator i to separator i\

Model

It will be assumed that the objective function can be expressed as a linear function that depends on the total flow to each separator. The model expressed in terms of concentrations and total flows has the following form:

min •= $\sum_{i=1}^{N-1} c_t S|$ (32.1)

st. reeo!•» $\sum_{t»i}^{N-l}$ *i * $\sum_{k*i}^{K}$ *m®k* IOZ.ZJ

L_{ij} s F | L_j **for all i and j** **(32.3)**

Si» F| + $\sum_{t'«l}^{1-1}$ RB,^{1} + $\sum_{!'«*•!}^{N-l}$ Kiy for aU i (32.4)

Si xsq = flj + $\sum^{1-1}$ RBf^1 x^j + $\sum^{N-l}$ KT$_r^!$ xtrj **for all i and j** **(32.5)**

S i s ^ + Bi **for alii** **(32.61**

T_i xt_{ij} = S_i xs_{ij} **for all i and ord(j) ≤ i** **(32.7)**

xt_{ij} = 0 **for aU i and ord(j) > i** **(32.8)**

B_i xb_{ij} = S_i xs_{ij} **for all i and ord(j) > i** **(32.9)**

xb_{ij} = 0 **for all i and ordQ) ^ i** **(32.10)**

T_f *m* $\sum_{1*1}^{t-i}$ RTi^r + $\sum_{k«l}^{K}$ FT$_t^k$ for aU i (32.11)

Bi s $\sum^{N-l}$ RB^^{f} + $\sum^{K}$ PB$_t^k$ **for all i** **(32.12)**

P^k = $\sum_{i=1}^{N-l}$ PT_i^k + $\sum_{i=1}^{N-l}$ PI3^ + 0^ Tor'alt k (32.13)

P_{kj} = $\sum_{i*l}^{N-l}$ PT,^{k} xt$_{ij}$ + $\sum_{i»l}^{N-l}$ PB,^{k} *xbq*+ a_k z, for all k and j (32.14)

$\sum_{ord(j)=1}^{i}$ **xt_{ij} =** 1 for alii (32.15)

$\sum_{ord(j)=i+1}^{N}$ **xb,j = 1** **for all i** **(32.16)**

$\sum_{ord(j)=i}^{N}$ **x s , j = l** **for alii** **(32.17)**

Si, T$_{|t}$ B,. Fi. RT/. RBi$^{*'}$. PT$_t^k$. PBi^k. c*. *t*^ xs,j, xty. xb_{ij} ≥ 0

The parameters Feed, z_j, P^k and p_{kj} represent the total feed, composition of the feed, total flow of product k and component flow of component j in product k, respectively. The variables xsq, xty and xbg are the concentrations of component j in the inlet stream to separator 1, top of separator 1 and bottom of separator i, respectively.

The objective function (32.1) is a linear function of the inlet flow to the separators. Equation (32.2) is the total mass balance in the inlet splitter and equation (32.3) is the component mass balance. Equations (32.4) and (32.5) are the total and component mass balances for the mixer i before the separator L The material balances for separator i are given by equations (32.6)* that is the total mass balance for the separator, equations (32.7) and (32.8) that are the component balances for the top stream and indicate that nothing from components number 1+1 to N-l is in the top of the separator, and equations (32.9) and (32.10) that are the component mass balances for the bottom streams. Equations (32.11) and (32.12) are the overall mass balances for the splitters of the top and bottom streams alter separator 1. The equations that state that the concentrations of the outlet streams should be the same that the inlet stream in a splitter have been already substituted. Finally, equations (32.13) and (32.14) are the overall and component mass balances for the mixer for product k.

Model (32) corresponds to a formulation of the type of model MX where the distribution coefficients are known and restricted to 0 or 1. Some simplifications have been made to avoid including many irrelevant variables (e.g. not to define concentrations for the streams that go the top i to product k). Although, some of the linear constraints in this formulation are redundant, they can become nonredundant in the linear relaxation as will be shown in Example 4.

Equations (32.5). (32.7). (32.9) and (32.14) involve nonconvex terms. This model can be reformulated as in model MR by introducing individual component flows and the linear equations (15M18) and (7) according to the approach illustrated earlier to obtain a model in the form of model MR The resulting reformulated model is as follows,

$$\min \Phi = \sum_{i=1}^{N-1} c_i S_i \qquad (33.1)$$

$$\text{st} \quad Feed = \sum_{i=1}^{N-1} F_i + \sum_{k=1}^{K} X_k \qquad (33.2)$$

$$f_{ij} = F_i z_j \qquad \text{for all i and j} \qquad (33.3)$$

$$S_i = F_i + \sum_{i'=1}^{i-1} RB_{i'i} + \sum_{i'=i+1}^{N-1} RT_{i'i} \qquad \text{for all i} \qquad (33.4)$$

$$s_{ij} = f_{ij} + \sum_{i'=1}^{i-1} rb_{i'ij} + \sum_{i'=i+1}^{N-1} rt_{i'ij} \qquad \text{for all i and j} \qquad (33.5)$$

Sr-TV+'B, for alii (33.6)

t_n = sij for all i and ord(jJ £ i (33.7)

t_f * 0 for all i and ord(j) > i (33.8)

$b_{,jSS}$Sg for all i and ord(j) > i (33.9)

t,j = O for all i and ordfl) £ i (33.10)

T, = $\sum_{1*1}^{t-}$ sW + $\sum_{k«l}^{K}$ lPT_4^k for all i (33.11)

B_i *m* $\sum_{i'=i+1}^{N-l}$ **£RB,^{if}** + $\sum_{k=1}^{K}$ £PB_t^k for all i (33.12)

p* = $\sum_{i=1}^{N-1}$ TF$T_t^{k''}$ + $\sum_{i=1}^{N-1}$ £PB|^k + otk for all k (33.13)

Pki * $\sum$ 5jtt,^{k} + $\sum$ Xpb^$^{k+}$ ctk z, for all k and j (33.14)

t« = $\sum_{i'»l}^{1*1}$ Z ^ / + $\sum_{k«l}^{K}$ Zptij^k foraUiandj (33.15)

b« = $\sum_{l'*t+l}^{N-l}$ Zrbij$^{4'}$ + $\sum_{k»l}^{K}$ £pb«^{k} for all i and j (33.16)

$\sum_{ord(j)=1}^{i}$ xt_{ij} = 1 for alii (33.17)

$\sum_{ord(j)=i+1}^{N}$ xb_{ij} = 1 for all i (33.18)

T_i = $\sum_{ord(j)=1}^{i}$ t_{ij} for alii (33.19)

B,s $\sum_{ord(j)=i+1}^{N}$ Ib« for alii (33.20)

PTi^k= $\sum_{oixi(j)»i'}^{i}$ pt<*^{k} for alii and k (33.21)

PB,^{k}= $\sum_{ord(j)=i+1}^{N}$ Ipbq^k for alii and k (33.22)

RT/ = $\sum_{ord(i)-l}^{i}$]DV for all i and i^f < i (33.23)

RB$_i^f$ = $\sum_{ord(j)=i+1}^{N}$ Irb/ for all i and r > i (33.24)

Equations (15-18) for t_{4J}, rt_{tJ}^l and pt_{tj}^k in terms of xt_{ij} and the total flow of its respective stream.

Equations (15-18) for bq, rbq* and pb_{fj}^k in tenns of xb_{ij} and the total flow of its respective stream.

S_i, T_{lt} Bt. F_4. *KTf*, RB,$^{1'}$, PT,^{k}. PB^. a_k. f,j. $xs_{,jf}$ $xt4_{jf}$ xb,j ^ 0

s_{ij}, t_{ij}, b_{ij} pt_{ij}^k, pb_j^k . rt,/. rby^{1}* ≥ 0

It is not necessary to include equations (15)-(18) for the inlet component flows to the separator, Sq. since the variables xsy only appear in these equations. Also, the component flows, S|j9 only appear in mixers and sharp separators units which can be exactly represented in terms of the individual component flow equations (33.5). (33.7) and (33.9). Equations (33.15)-(33,16) that are the component mass balances for the splitters of top and bottom streams have been included accordingly to the reformulation previously presented. Equations (33.19M33.24) relate the total flow and the individual component flows for the splitter streams.

Preprocessing

The proposed Superstructure (Fig. 9) allows to bypass certain amount of the feed to the product k. Ok, without having to go through the separation network. The amount of the product k that is not bypassed has to be processed In the separation network and it will be denoted as the 'residual product*. Hence, the total 'residual product k^f is given by (P^k - a_k) and has the component flows given by (p^ - o^Zj)(see Fig. 10).

The global optimal solution of model (32) is a network in which all the 'residual products[1], have at least one component with a zero flow. The reason that it is not optimal to separate a stream in the network and later to remix it. The same degree of separation can be achieved using a bypass that does not incur any cost in the objective function.

Consider the second separator in the solution obtained by Floudas (1987) to his second example (see Fig. 11). For this subnetwork of the complete structure the "upper 'residual product' has components B and C present. The components are being separated and remixed again. The same outlet flows can be obtained with a smaller input flow to the separator as it is shown in Fig. 11. Note that both 'residual products' have components with zero flow.

It should be clear that if there was not a component with zero flow in the 'residual product[1], then there is part of this stream that could have been obtained by just bypassing the network. This in turn does not incur in any cost, whereas going through the network has a positive cost. The above condition gives a lower bound for the bypass to each product. This also corresponds to the largest amount that can be bypassed since all this flows in the 'residual flow[1] have to be positive. In this form the bypass can be precalculated without affecting the global optimality of the solution.

The bypass to product k is given by the maximum amount that can be sent to product k without having a negative flow; that is.

$$a^* = \min_j t \wedge 4 \qquad (34)$$

where Zjis the feed composition and p_{kJ} is the flow of component j in product k. The component flews for 'residual product[1] k, *y#* are given by.

$$\gamma_{kj} = p_{kj} - \alpha_k z_j \qquad (35)$$

Key component bounds

Wehe and Westerberg (1987) proposed using lower bounds for the flow of the key components in separator i. These bounds are based on the fact that separator i is the only unit that can perform the task of separating component number i from component nutaber i+1. They are redundant for the nonlinear formulation (32) but they are relevant for the linear relaxation in (33). To calculate them, it is necessary to determine in each product what is the difference between the two key components of separator 1 with respect to the concentrations in the feed. The lower bounds in separator i* for the flow of the key components in the top (component Jl) and bottom (component j2) streams are given by:

t|i * I {IWj - zji nitowi J2 nn) (k=1; γki/zj) for all i (36)

$$b_{ij2} \geq \sum_{k=1}^{K} \{ \gamma_{kj2} - z_{j2} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (37)$$

where *y^* is given by (35). It is important to include both bounds in the relax model (33) since there is no guarantee that the inlet flow to separator i has the same proportion between the key components as the feed. It is not known in which part of the sequence separator 1 will be placed, and it can be after a splitter that is not being approximated correctly.

The bounds in (36) and (37) can be extended to separation of components that are not adjacent in the feed. Consider component number i and component number i+3. There are three separators that can perform this task, separators i. i+1 and i+2. Cuts of the following form can be obtained,

t* i + tf! t + t,«a *i Z* I {Tkt - z, mtaonHH or *3 l~Tl) (K; k=1; γkj) for all i (38)

$$b_{ii} + b_{i+1\,i} + b_{i+2\,i} \geq \sum_{k=1}^{K} \{ \gamma_{ki+3} - z_{i+3} \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (39)$$

Equations as the ones in (38) and (39) can be redundant compared to equations (36) and (37) and it is possible to detect this before solving the problem.

Relativeflowrate constraints

These constraints are used when the relation between the flowrates of two components is known. In particular, consider component A in the last column of the network (we Fig. 12). None of the redirected streams contains component A. Therefore, the relative flowrate of component A with respect to the other components in the top stream has to be smaller than in the feed. This relation should remain valid after the top stream is split to the products and redirected flows.

In the separator previous to the last one, N-2, all the streams do not have component A except the one coming from the top of the last column. For this one it is already known that the relative flow of component A with respect to the other components is smaller than in the feed. This type of analysis can be done for component A and component N in all the columns yielding th$ following linear constraints for the splitters.

for alii and k

$ZAPtij^{k}$-ZjPt«A^{k}^O	forj *Aandord(j)≤i	(40)
z_N pb,j^{k} - z, pt*$_N^{k}$ £ 0	for ord(j) * N and ord(j) > i	(41)

foralliandi'>i

ZArt|j^{r}•ZjrtiA$^{4'}$^0	for J * A and ord W ^ *	W2)

for alii and f<i

z_N rb$_{ij}^{r}$ - z_j rb$_{iN}^{r}$ ≥ O	forord(J)*Wandord(j)>i	I43J

Boundson concentrationsand total flows

The approximations (15-18) require bounds for the total flows and component concentrations in the splitters. The lower bound for the total flow of the top and bottom streams is given by the lower bound of the key components obtained in equations (36) and (37). For the outlet streams of the splittters, that are the redirected streams and the streams that go to the products, the lower bound is zero. The upper bound for the total flow of the top and bottom streams is given by the feed to the network of the components that can be present in each stream .that is,

$$T_t^u = \sum_{ord(j)=1}^{t} Z(Feed-Xak)z, \qquad \text{for ail i} \qquad (44)$$

$$B|« = \sum_{ord(j)=i+1}^{N} ItFeed-IOfcJz, \qquad \text{forallj} \qquad (45)$$

The upper bound for the streams after the splitter are given by.

$$RT_i^{i'u} = T_i^u \quad \text{for all i and i'} < i \quad (46)$$

$$RB_i^{i'u} = B_i^u \quad \text{for all i and i'} > i \quad (47)$$

$$PT_i^k = \sum_{ord(j)=1}^{i} \gamma_{kj} \quad \text{for all i and } k \quad (48)$$

$$PB_i^k = \sum_{ord(j)=i+1}^{N} \gamma_{kj} \quad \text{for all i and k} \quad (49)$$

The lower bounds for the concentrations are zero except for the key components in the separator for which the lower bounds are given by the lower bound of its flow divided by the upper bound of the total flow of that stream. The upper bounds in the concentrations are given by one minus the lower bounds of the other components.

The solution of the linear programming model (33) provides a lower bound to the global optimum since this model is a valid relaxation of the nonconvex model (32). This lower bound is obtained by solving the LP model for the 'residual products' in 135) with the addition of the valid constraints (36)-(43).

The upper bounds are generated solving mode} (32) in terms of the individual flows for the 'residual products'. When additional nonconvex problems are solved to improve the current upper bound it can happen that very similar initial point are generated. In this case, a new nonconvex NLP is solved in which bounds over the total inlet flows to the separators (SJ are included. For this purpose the values of these variables in the LP solution (S_4*) are used such that the current incumbent solution is no longer feasible.

Example4

Consider the 3 component example proposed by Floudas and Agganval (1990). An equimolar feed has to be separated into two products as show in Fig. 13. The objective function is given by

$$\Phi = 0.2395 + 0.00432\ S, + 0.7584 + 0.01517\ \&* \quad (50)$$

The bypass to products 1 and 2 can be calculated according to equation (34) and the 'residual product' component flows are obtained through equation (35) (see Fig. 14). The total bypass to product 1 is a, = 90 and the bypass to product 2 is ct_2 = ISO and the feed has a concentration of z_A = 1/3, z_Q = 1/3 and Zc = 1/3. In this form the 'residual product' 1 is YIA = 0, YiB = 20 and y_{lc} = 0 and the residual product' 2 is Y_2A = 20, Y_{2B} = 0 and Y2C = 20. Additionally, lower bounds on the flow of the key components in both separators are obtained

using equations (35)-(36). The key components in separator 1 are component A in the top and its flow has to be at least 20 and component B in the bottom has to have at least a flow of 20. In the top stream of the second separator at least 20 units of component B have to be separated from 20 units of component C in the bottom stream. It is important to note that after preprocessing the network several suboptimal solutions have been cut off. One of these suboptimal solutions for this particular data is a parallel configuration of both separators (there are situations in which a parallel configuration can correspond to the global solution as will be shown in example 5). In this example the direct or indirect sequence have a lower objective function. Both of these configurations are local solutions with an objective function value of 4 » 1.8639 for the direct sequence and 4 = 2.081 for the indirect one. In some instances, MINOS 5.2 had problems converging even in this small example.

The LP (33) is formulated for this problem, giving a lower bound of (^ = 1.8639. The approximations are exact and therefore this solution is a feasible solution of model (32) proving that it corresponds to the global optimum. Hence, convergence is achieved in one iteration. The optimum solution corresponds to the direct sequence shown in Fig. 15. It should be noted that if the linear mass balances for the mixer for product 2 were not considered since they are redundant for the nonlinear formulation, a lower bound in the relaxed model of Φ_L = 1.12 is obtained, this shows that it is relevant to include all the mass balances in the linear model in order to tighten the lower bound.

Example 5

In the approach proposed by Wehe and Westerberg (1987) for the case of 3 components only the direct and indirect sequences are considered and both options can be modelled as LP problems since no mixing is required for these separation networks. However, this example shows that parallel configurations can be also globally optimal and that they are not excluded by the method proposed in this paper. To be able to consider parallel configurations or any combination of parallel with direct or indirect sequences it is necessary to model a superstructure in which mixing is allowed (like in the structure used in Fig. 13). Here, nonconvexities arise in the mass balance equations after the separators.

Consider that an equimolar feed is to be separated into the two different products given in Fig. 16. The objective function is to minimize the sum of the total flows into the separators. The same procedure that in the previous example is followed and the bypass can be precalculated by equation (34). The solution to the model (32) yields Φ_L = 12 and since it is a feasible solution of model (32) it corresponds to the global optimum (see Fig. 16). Note that the solutions for the direct or indirect sequences have an objective function of Φ =16.

Branch and Bound

If there is a gap between the lower and upper bound a branch and bound search is performed. It is only necessary to do the search over the variables involved in the nonconvex terms. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter* In this way. it is necessary to check the approximation for the concentrations in the splitters of the top and bottom streains of the separator. Equations (26)-(29) for the splitters of top and bottom streams are used to perfom the branch and bound search.

Results

Table 1 summarizes the results of the earlier examples 1 to 3 and of the sharp separation network examples 4 to 12. The number of variables is the total number of variables that are included in the reformulated and relaxed model (33) for that example. The lower bound is the initial bound that *fs* obtained by solving model (33) over the entire feasible space. The inital gap represents the percentage difference between the initial lower and upper bounds. When there is a zero initial gap it means that the first relaxed solution is feasible in the original problem thereby corresponding to the global solution. The column for nodes gives the total number of subproblems that where solved before converging to the global solution. A relative tolerance of 0.01 was used, except for example 2 were exact convergence was obtained after branching and example 12 for which a tolerance of 0.02 was used. It is important to note that the initial lower bound is tight and that it corresponds to a good estimation of the global solution. The largest differences are for example 1 with a 25% of difference and for example 12 with a 7% difference. The LP time refers to the time used to solve each relaxed model and the NLP time is the time used for solving a nonconvex model. It is possible to do updates using the previous LP solution and in this form have a more efficient implementation. The times are in seconds and the problems were solved on an IBM RS600/530 using GAMS 2.25 (Brooke et al. (1988)). MINOS 5.2 was used to solve the LP problems and CONOPT for the nonconvex NLP problems. A brief description of the example problems 6 to 12 is given below. It includes the specific data for the problem, the objective function and the topology of the network that is the global solution.

Example 6

This example corresponds to example 2 from Floudas (1987). In this case a linear objective function with the same cost coefficients is used and it is given by,

$$\Phi = 2.5\, S_1 + 3.0\, S_2 + 1.5\, S_3 \qquad (51)$$

The data for the composition of the products is given in Table 2.

The initial lower bound is Φ_L = 54.25 and an upper bound of 0 = 55.5 is obtained by solving the nonconvex problem. A partition of the feasible region is performed using the composition of component D in the bottom stream of separator 1. The first subproblem (x £ 0.166) yields a lower bound of Φ_L = 55.45 and the second subproblem (x ≤ 0.166) has a solution of Φ_L = 55.8. The latter is greater than the upper bound and the former is less than 1% of the global solution (see Fig. 17).

Exajnple7

This example is taken from Floudas (1987). The data for this problem is given in Table 3 and the linear objective function is given by:

$$\Phi = 2.5S_1+3.0S_2+L2S3 \quad (52)$$

The initial lower bound is Φ_L = 32.7 and it provides a feasible solution to the nonconvex problem. In this form the global solution (see Fig. 18) is obtained in one iteration. It is interesting to see that this solution also provides a better objective function for the concave objective function used by Floudas (1987) ($ = 10.65 versus $ = 13.68 which is 28% higher)

Example 8

Tills four component problem is taken from Wehe and Westerberg (1987). The data for the products is given in Table 4 and the objective function has the following form:

$$\Phi = 5.0 + 0.5\ Si + 4.0 + 0.3\ S2 + 6.0 + 0.7\ S_3 \quad (53)$$

The first relaxed subproblem has a solution of 0_L =26.76 and it is infeasible for the nonconvex problem. A nonconvex problem is solved using CONOPT with the LP solution as the initial point. An upper bound of 4> = 26.79 Is obtained corresponding to the global solution (see Fig. 19) within a 0.1%.

Example 9

This example corresponds to example 1 from Wehe and Westerberg (1987). Table 5 provides the data for the product flows and the objective function is given by:

$$\Phi = 5.0 + 0.5\ Si + 9.0 + 1.0\ S2 + 3.0 + 0.4\ S_3 + 6.0 + 0.6\ S_4 \quad (54)$$

A initial lower bound of Φ_L = 85.16 is obtained and the upper bound Is $ « 85.65. The difference is 0,5% and the global solution (see Fig. 20) is obtained in one iteration.

Example 10

This problem is taken from Floudas (1987) and the data is given in Table 6. The objective function is given by,

$$4 \ll 1.2\ Sj + 3.0\ Sj + 2.5\ S_3 + 1.5\ S_4 \qquad (55)$$

The JrHtui lower bound is <^ = 156.56 and the upper bound is $ = 179.08. After 5 nodes the global solution of $ = 159.48 (see Fig. 21) is obtained.

Example 11

The data for this 6 component problem are given in Table 7 and the objective function has the following form:

$$4 \gg i.5\ S, + 3.0\ Sj + 2.0\ S3 + I.OS4 + 4.0\ S_5 \quad . \qquad (56)$$

The initial lower bound is 0_L = 173 and the upper bound is 4 » 179.11. After five nodes the global solution is obtained (see Fig. 22).

Example 12

This is a 6 component 4 products problem and the data are given in Table 8. The objective function is:

$$\$\ m\ 5.0\ Si + 3.0\ S2 + 2.0\ S_3 + 2.5\ S_4 + 4.0\ S_5 \qquad (57)$$

The initial lower bound is ϕ_L = 362 and the initial upper bound is 0 = 415.6. The global solution of 4> = 388 (with a 2% optimality gap) is obtained after 33 nodes (see Fig. 23).

Conclusions

A general procedure for the global optimization of process networks with multicomponent streams has been proposed. The basic idea relies on a relaxed LP model that is obtained through reformulation-linearization techniques that establish a clear relation between the component flow and the composition models for mass balances. The reformulated model combines both of these providing tighter lower bounds than other relaxations proposed in the previous work. The relaxed linear model has been embedded in a branch and bound procedure to obtain the global optimal solution.

As has also been shown, the algorithm can be further specialized to take advantage of the particular structure of sharp separation networks with single feed and mixed products. Here, it is possible to preprocess the problem to reduce the space over which the search is

conducted. The bounds that are necessary for the estimator functions in the relaxed model can be obtained without having to solve any subproblems. Different types of linear approximations that are nonredundant to the relaxed model are included to obtain a tighter lower bound.

Twelve examples for both general process networks and for sharp separation networks have been presented to illustrate the performance of the algorithm. As has been shown, only a small number of nodes are commonly needed in the branch and bound search to identify the global or e-global solution. Moreover, in many cases the initial lower bound is either the exact solution or a very good approximation to the global solution.

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Appendix A. Reformulation-Linearization to obtain the nonlinear constraints in model MF

The nonlinear equations in model MF. that can be expressed either as (8) or (9), can also be generated from model MX. For this purpose take the concentration model MX and consider equation (5),

$$x_j^k = x_j^l \qquad \text{(A.1)}$$

multiply by the valid bound constraint X_j^k £ 0

xfvfm^xf (A.2)

Use equation (5) for component J\

xfxf**xtXf^k (A.3)

Multiply by the valid bound constraints $F^k \geq 0$ and $F^1 \geq 0$,

$F^k x,^k F^l X f^4 = F*\wedge^l F^k x \wedge^k$ (A.4)

that it is linearized to yield.

$$f_j^k f_j^l = f_j^l f_j^k \qquad \text{(A.5)}$$

which is precisely equation (8) for the splitters in the individual flow model MF.

Consider again equation (5),

$x,^k = \wedge$ IA.6)

multiply by the valid bound constraints $F^k \geq 0$ and $F^1 \geq 0$,

F^xfF^F'xj'F^1' IA.7)

that can be linearized to yield.

$$f_j^k F^l = f_j^l F^k \qquad \text{(A.8)}$$

Define the split fraction £ to be,

? » £ (A.9)

Equation (A.8) can then be expressed as

fjt^tff (A. 10)

which corresponds to equation (9).

Hence, the nonlinear equations (8) and (9) are redundant to model MX. Their linear approximations in general are also redundant in the linear reformulated model MR. Consider equation (A. 10), similarly to (15) one of its linear approximations is given by.

$$f_j^l \geq \xi^{lL} f_j^k + \xi^l f_j^{kL} - \xi^{lL} f_j^{kL} \qquad \text{(A.11)}$$

If there are no particular restrictions in the splitters, then the bounds for the split fraction variable are 0 £ £^{f} ≤ 1 and using them in (A. 11) yields.

$$f_j^1 \geq f_j^{kL} \xi^1 \qquad \text{(A.12)}$$

The bound for the individual component flow is given by $f_j^{kL} = x_j^{kL} F^{kL}$; also $x_j^k = x_j^1$ and £^{f} = $\frac{F^1}{p£_f}$ which leads to,

$$f_j^1 \geq x_j^{1L} F^1 \frac{F^{kL}}{F^k} \qquad \text{(A. 13)}$$

The estimator (15) for the same conditions ($F^{1L} = 0$) is given by

$$f_j^1 \geq x_j^{1L} F^1 \qquad \text{(A.14)}$$

Since the factor $\frac{F^{kL}}{F^k}$ is always less or equal than 1. equation (A. 13) is redundant. A similar analisis can be performed for the other estimators. Only when more specific bounds over the split fractions or the individual component flows are known, will these additional estimators be non redundant.

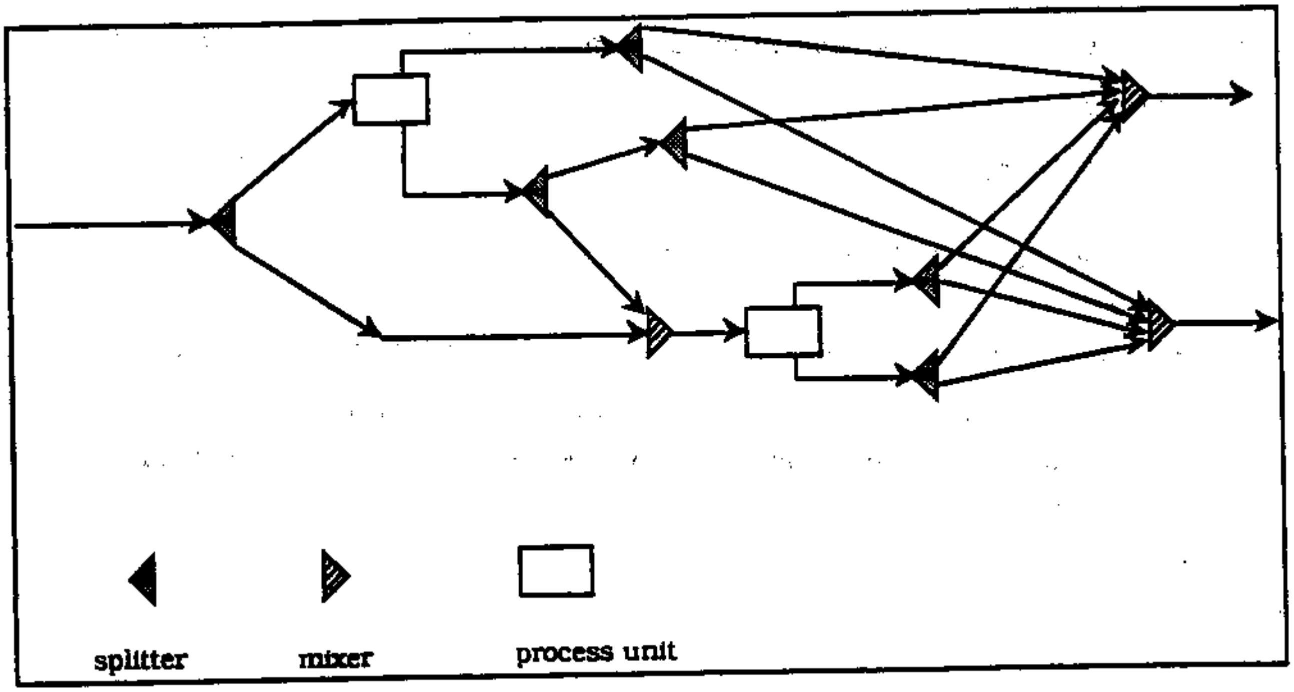


Figure 1. Process network with units, splitters and mixers.

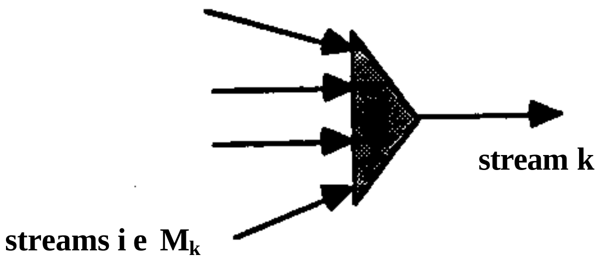


Figure 2. Mixer module.

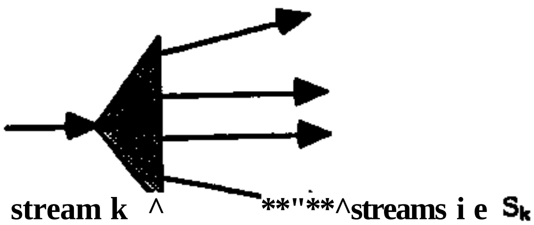


Figure 3. Splitter module.

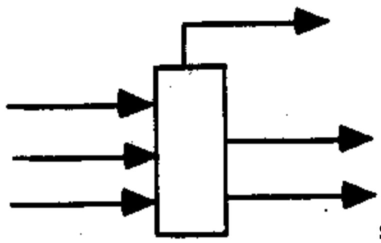
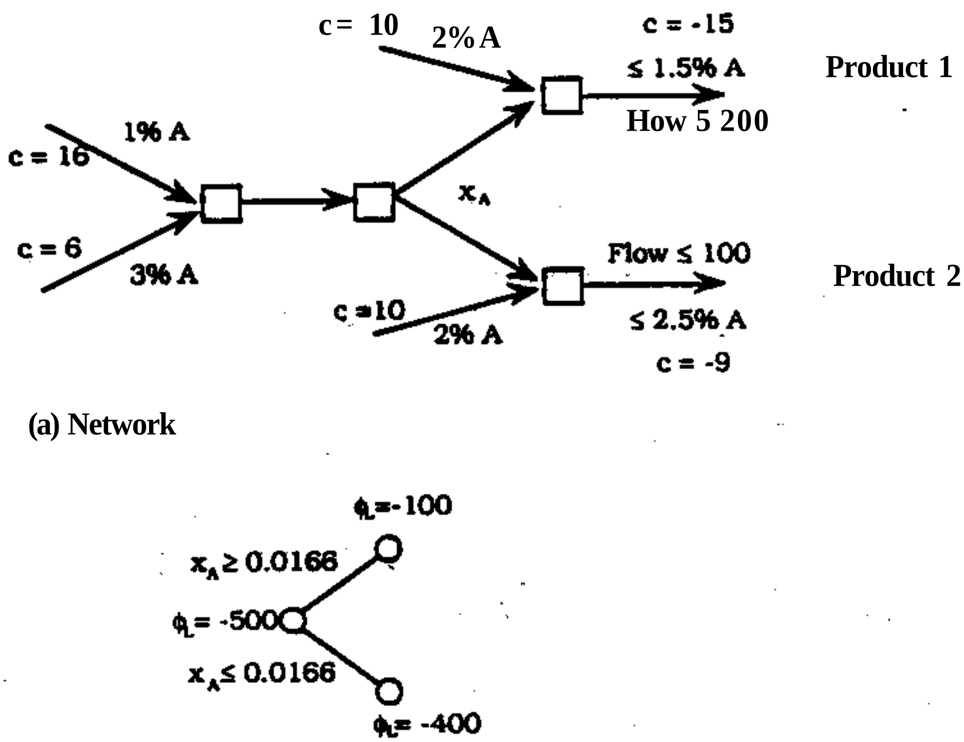


Figure 4. Process unit module.



(a) Network

(b) Tree search

Figure 5. Network and branch and bound search for example 1

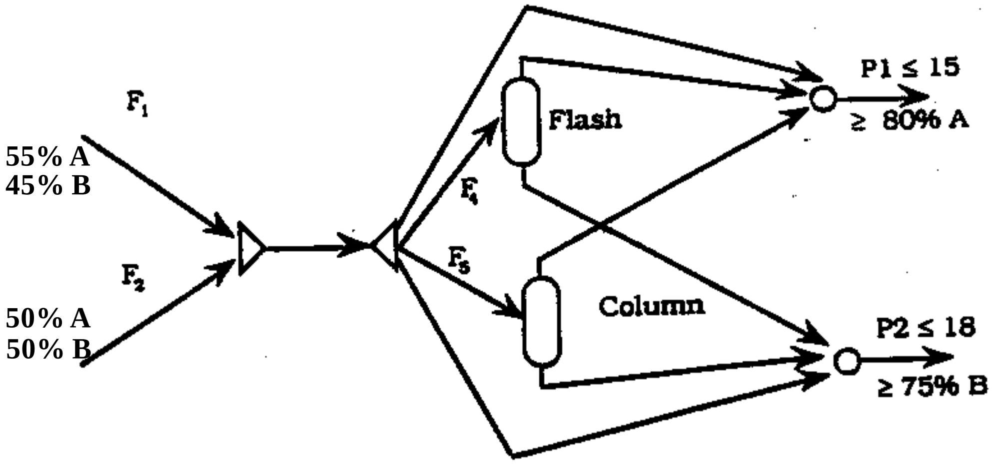


Figure 6. Network for example 2.

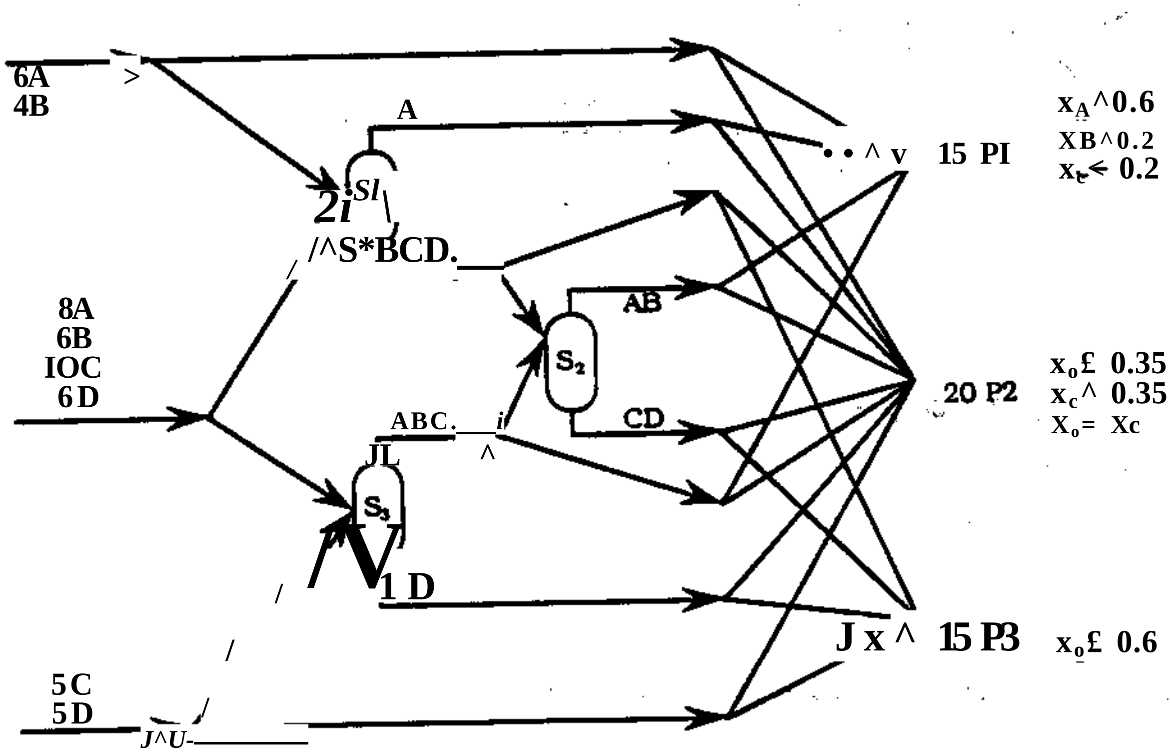


Figure 7. Network for example 3.

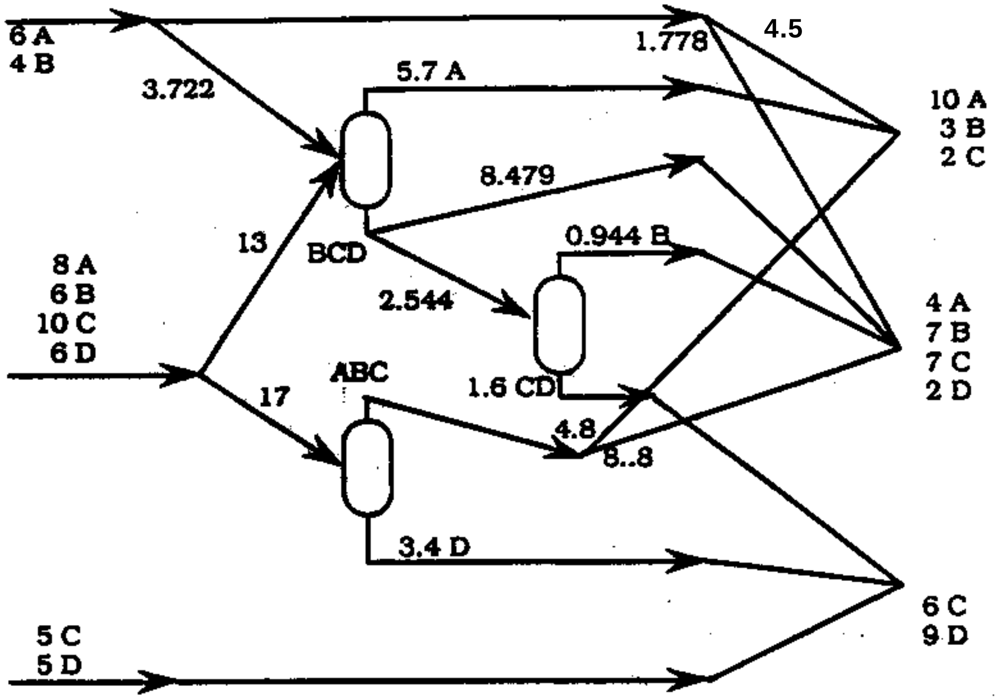


Figure 8. Optimal network for example 3.

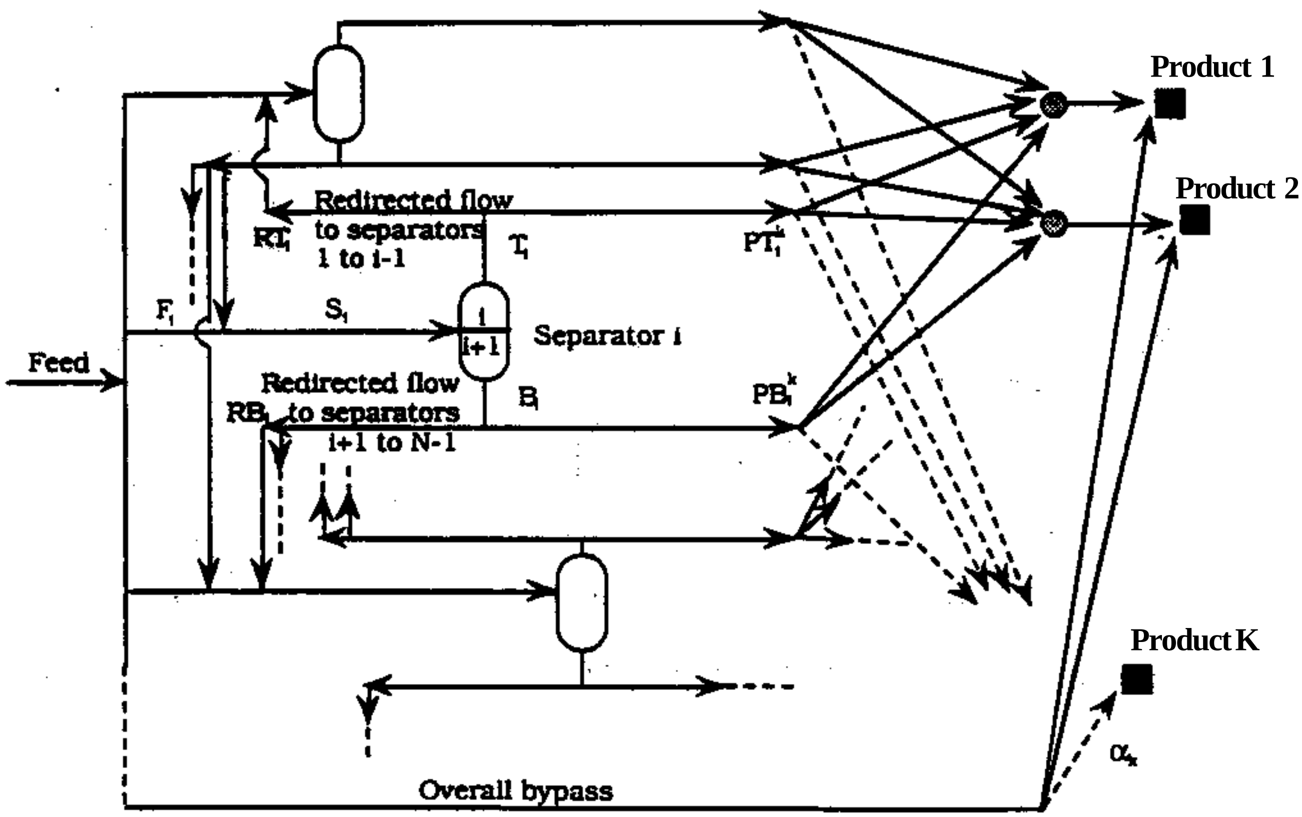


Figure 9. Superstructure for separation with sharp splits and mixed products.

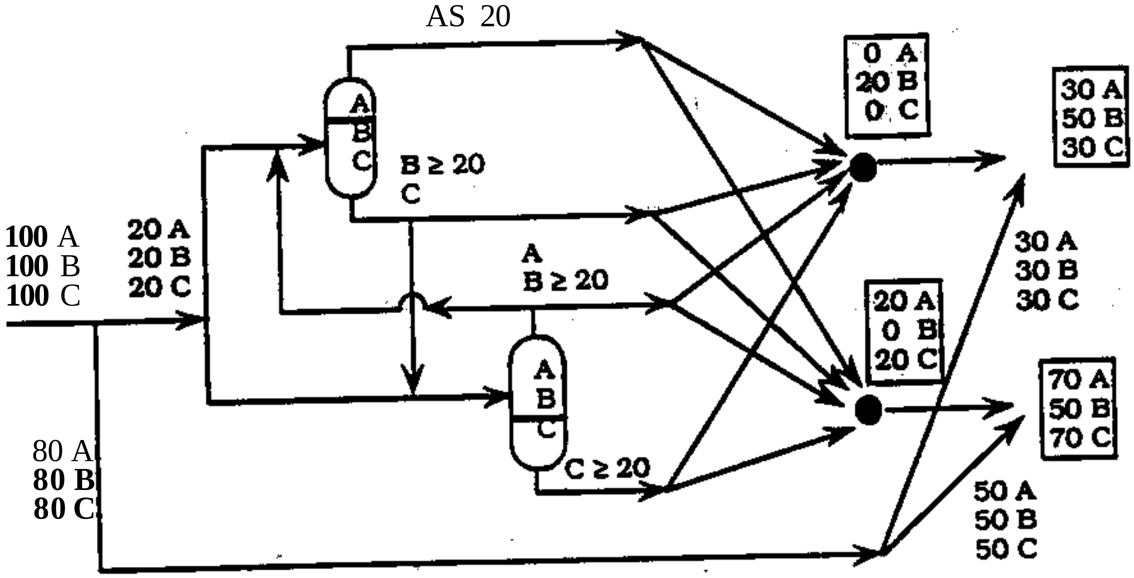


Figure 14. Residual products and key component bounds in example 4.

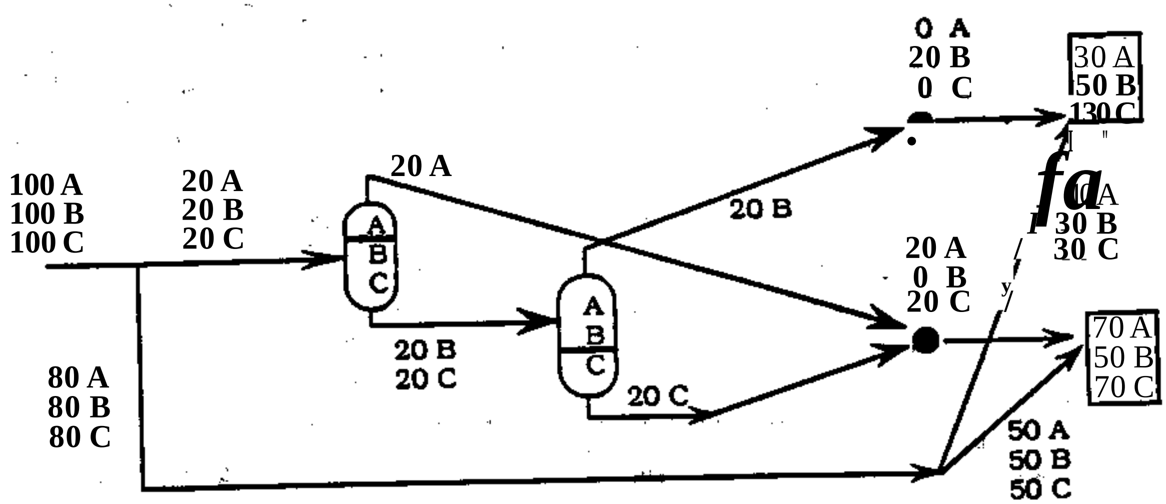


Figure 15- **Global** optimum solution of example 4.

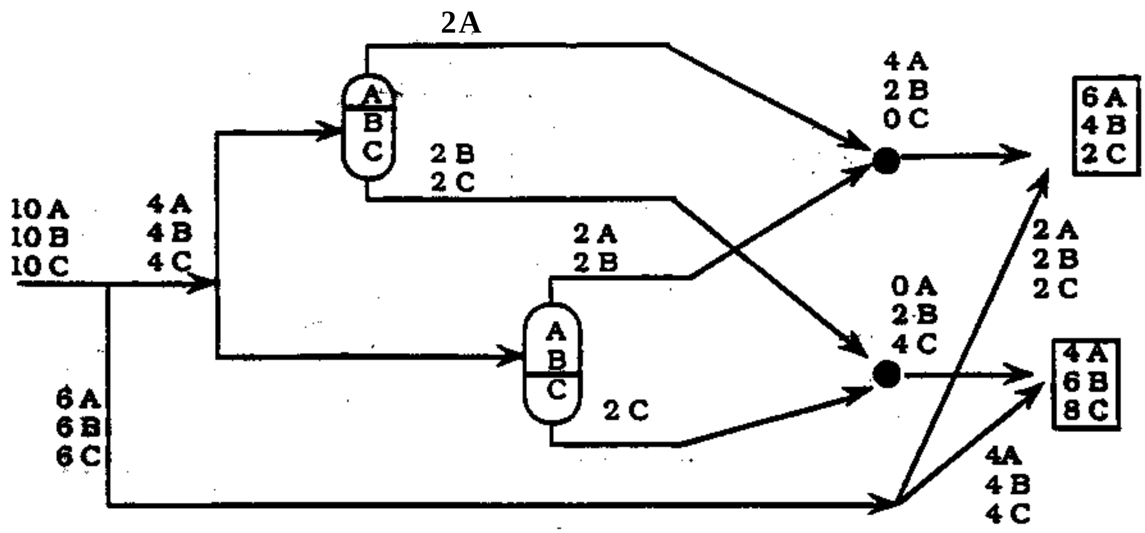


Figure 16. Global optimum solution of example 5.

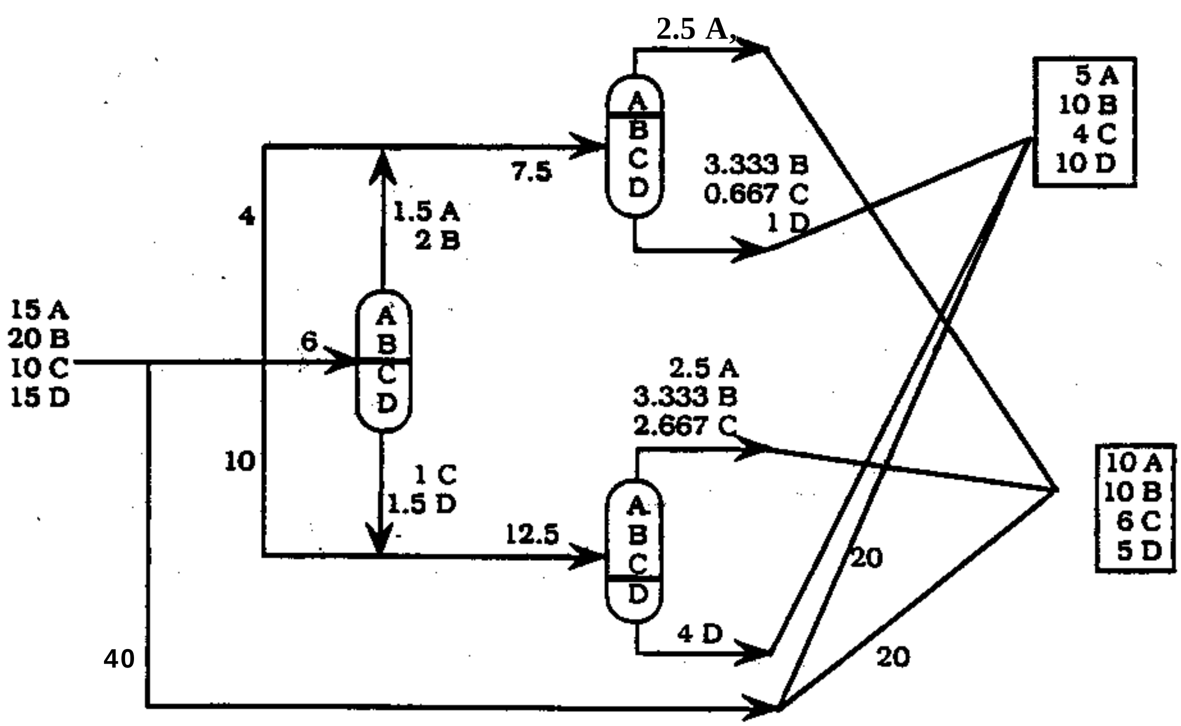


Figure 17. Solution of example 6.

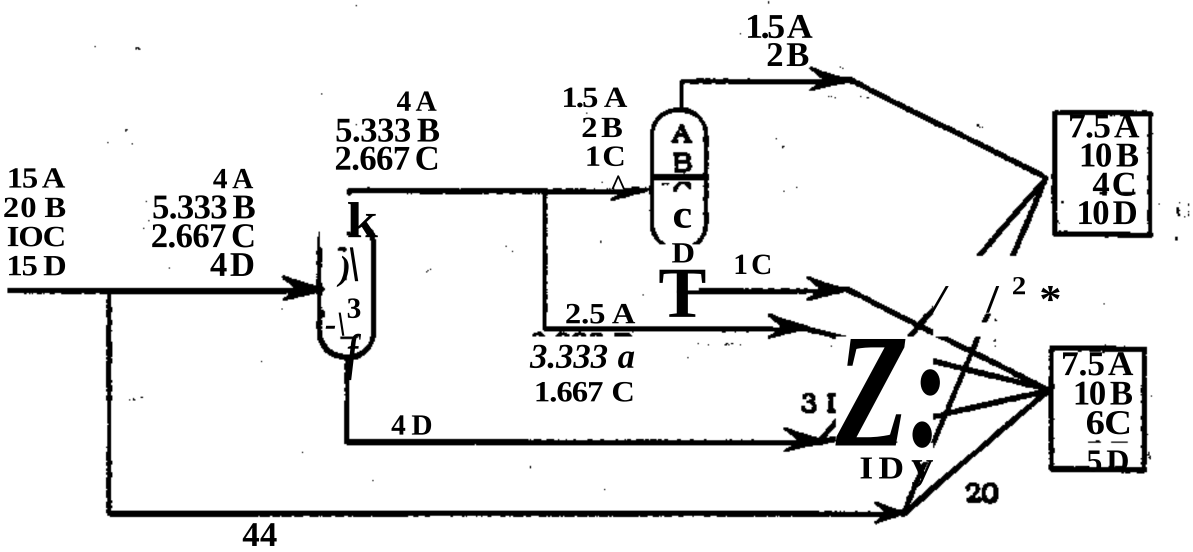


Figure 18. Solution of example 7.

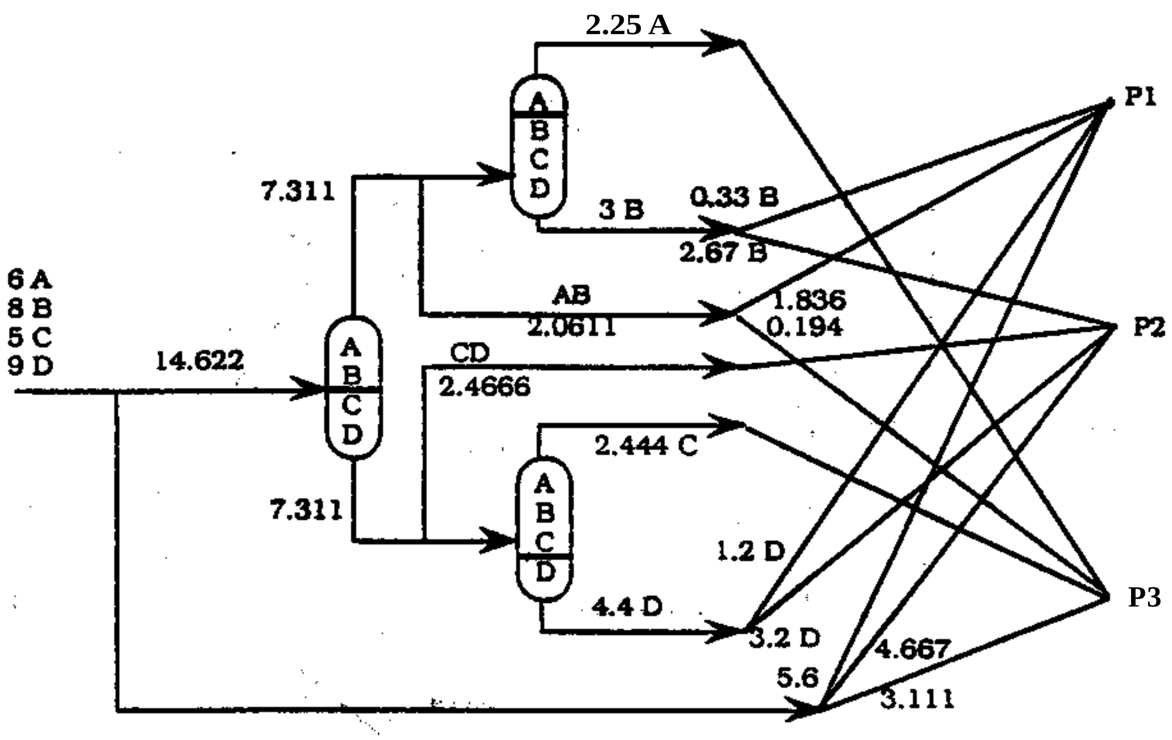


Figure 19. Solution of example 8.

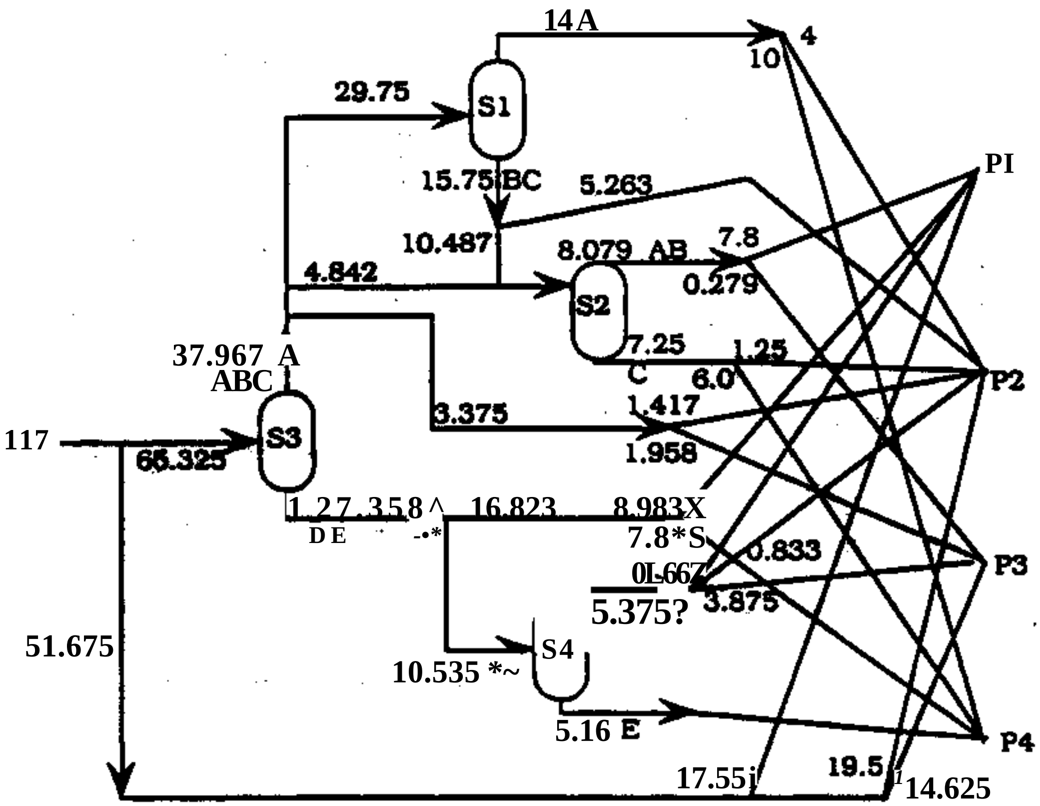


Figure 20. Solution of example 9.

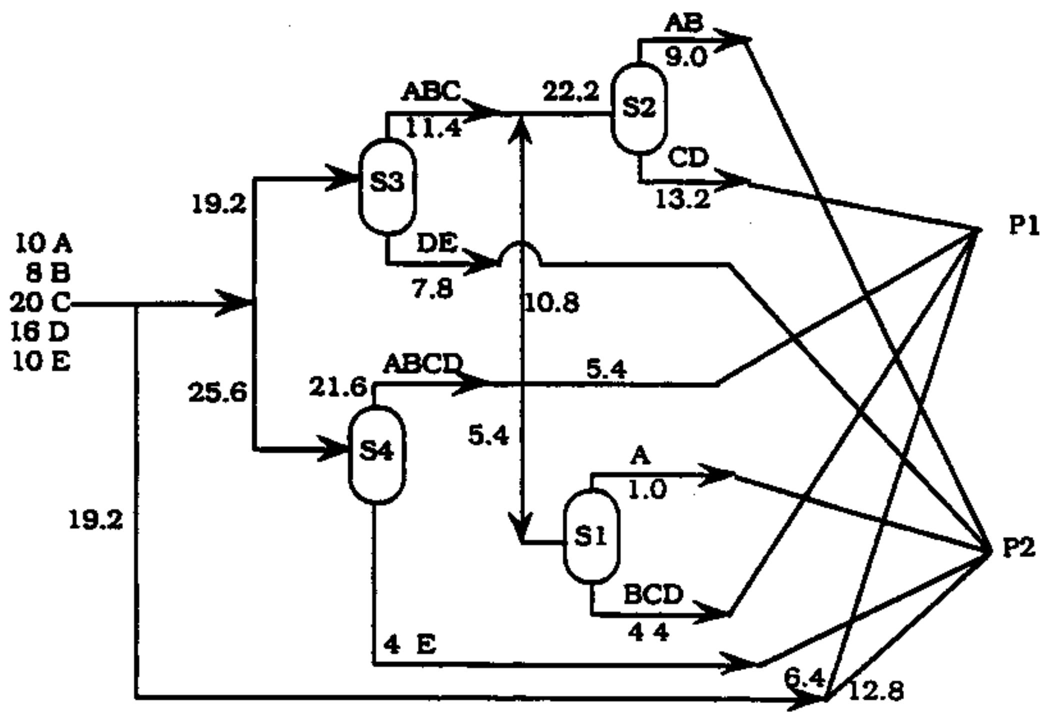


Figure 21. Solution of example 10.

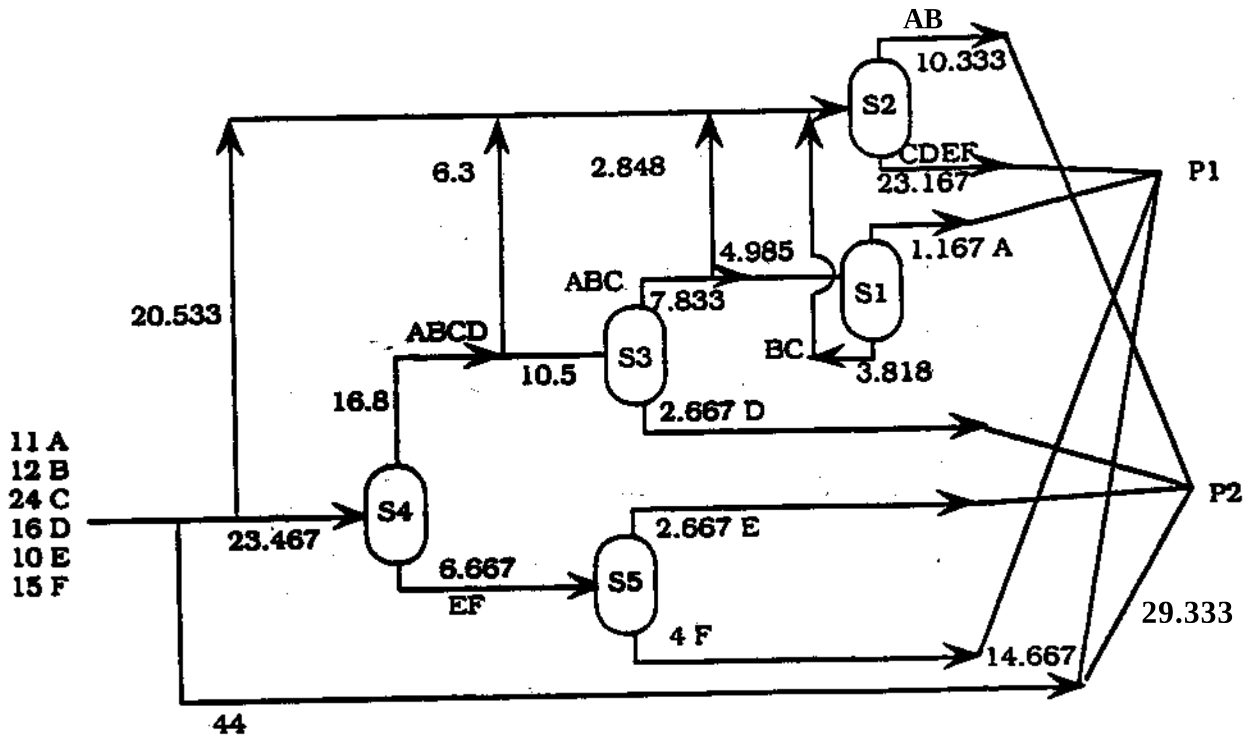


Figure 22. Solution of example 11.

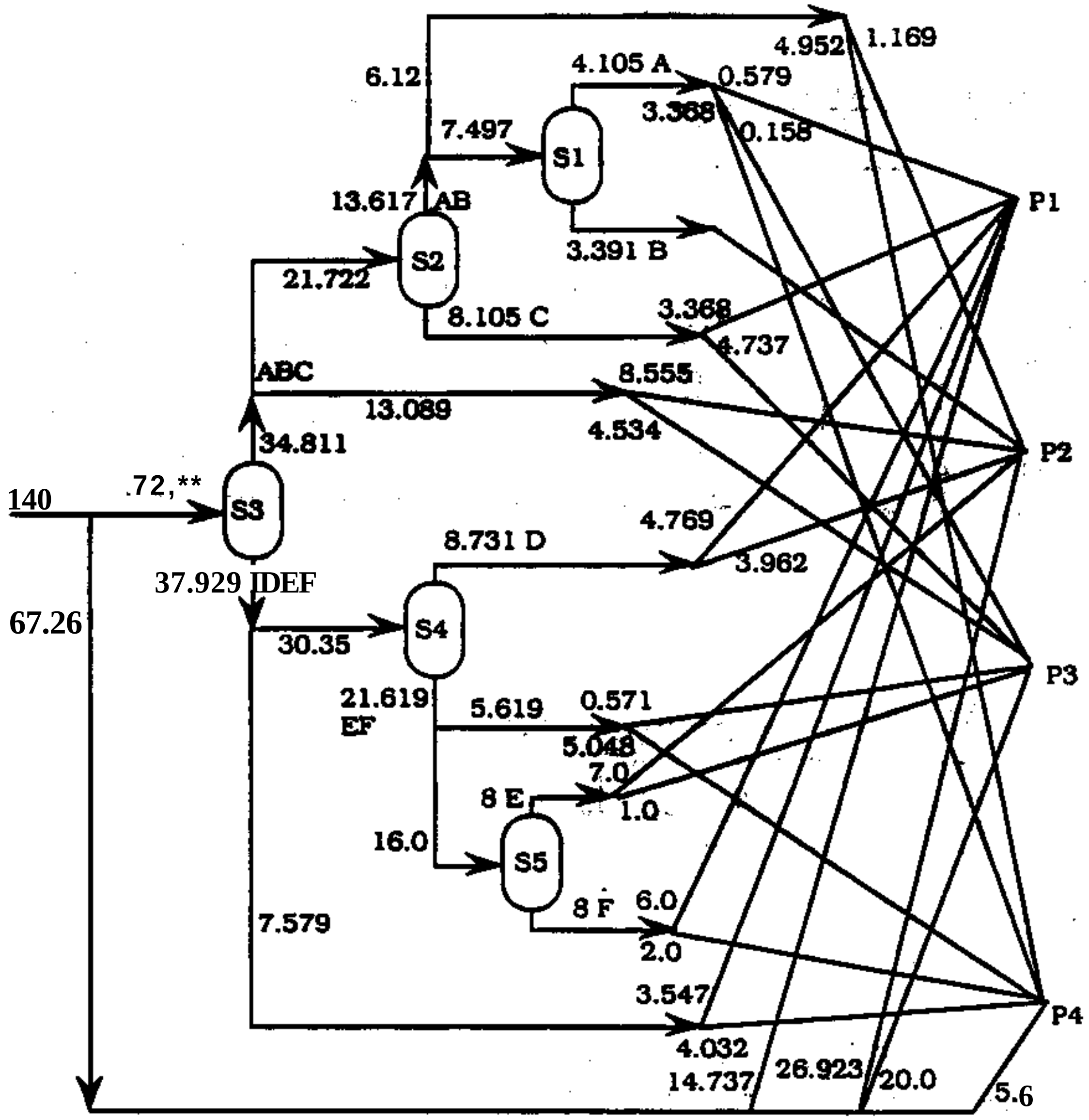


Figure 23. Solution of example 12.

Table 1. Computational results.

	Comp.	WocL	Var.	Lower bound	Initial gap	Global solution	Nodes	LP time	NLP time
Example 1	—	...	29	-500	20'	-400	3	0.05	0.1
Example 2	—	...	35	-513.22	0.3	-511.87	3	0.26	0.3
Example 3			113	138.18	0.4	138.7	1	0.34	0.4
Example 4	3	2	65	1.8639	0.0	1.8639	1	0.13	-.
Example 5	3	2	65	16	0.0	16	1	0.13	..
Example 6	4	2	107	54.25	2.3	55.5	3	0.97	0.4
Example 7	4	2	107	32.7	0.0	32.7	1	0.17	..
Example 8	4	3	125	26.76	0.1	26.79	1	0.23	0.3
Example 9	5	4	281	85.16	0.5	85.65	1	3.08	2.8
Example 10	5	2	225	156.56	12.4	159.48	5	2.59	2.3
Example 11	6	2	350	173	3.5	179.11	5	9.98	8.8
Example 12	6	4	430	362	14.8	388	33	19.8	13.2

Table 2. Data for example 6.

Component	A	B	C	D"	Total
Product 1	5	10	4	10	29
Product 2	10	10	6	5	31
Feed	15	20	10	15	60

Table 3. Data for example 7.

Component	A	B	C	D	Total
Product 1	7.5	10		10	31.5
Product 2	7.5	10	6	5	28.5
Feed	15	20	10	15	60

Table 4. Data for example 8.

Component	A	B	C	D	Total
Product 1	2	3	1	3	9
Product 2	1	4	1	5	11
Product 3	3	1	3	1	8
Feed	6	8	5	9	28

Table 5. Data for example 9.

Component	A	B	C	D	E	Total
Product 1	7	8	3	9	8	35
Product 2	10	3	5	5	4	27
Product 3	5	5	6	7	3	26
Product 4	10	0	6	4	9	29
Feed	32	16	20	25	24	117

Table 6, Data for example 10,

Component	A	B	C	D	E	Total
Product 1	2	2.4	16	8	1	29.4
Product 2	8	5.6	4	8	9	34.6
Feed	10	8	20	16	10	64

Table 7. Data for example 11.

Component	A	B	C	D	E	F	Total
Product 1	3	2	16	8	4	10	4$
Product 2	8	10	8	8	6	5	4$
Feed	11	12	24	16	10	15	88

Table 8. Data for example 12.

Component	A	B	C	D	E	F	Total
Product 1	3	2	6	8	4	10	33
Product 2	8	10	8	8	6	5	45
Product 3	5	4	10	3	11	4	37
Product 4	7	3	1	2	5	7	25
Feed	23	19	25	21	26	26	140

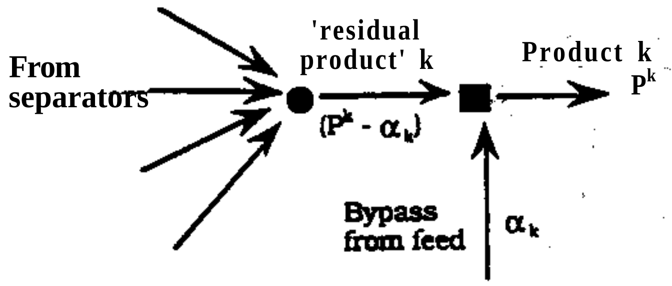


Figure 10. Definition of'residual product'.

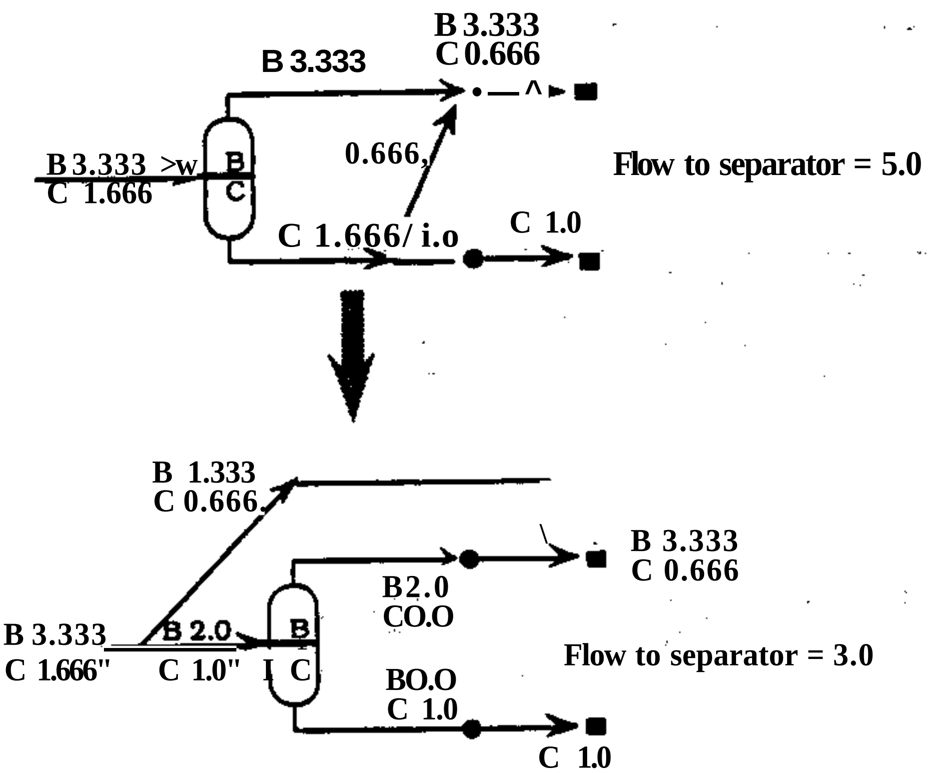


Figure 11. Example of solution without and with a zero component flow in 'residual product".

Global Optimization of Process Networks with Multicomponent Flows

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Abstract

This paper deals with the global optimization of networks consisting of splitters, mixers and linear process units and which involve multicomponent streams. Examples include pooling and blending systems and sharp separation networks. A reformulation-linearization technique is first applied to concentration and flow based models in order to obtain a relaxed LP formulation that provides a valid lower bound to the global optimum. This formulation is then used within a spatial branch and bound search. The application of this method is considered in detail for sharp separation systems with single feed and mixed products. Numerical results are presented on twelve test problems to show that only few nodes are commonly required in the branch and bound search.

Introduction

A common source of nonconvexities in the synthesis and design of processes, as well as in flowsheet optimization, are the material flow equations for multicomponent streams. These nonconvex equations involve bilinear terms and they arise in the mass balance equations when the compositions are unknown. There are different equivalent formulations for this type of networks. One alternative is to formulate the mass balance equations in terms of component concentrations* In this form bilinear terms are present in the equations for the mixer units and the different process units (e.g. sharp separators). A second alternative is to express the mass balances in terms of flows of individual components. This option has the advantage that it involves a smaller number of nonlinear equations. However, the modelling of the splitter units involves bilinear terms that arise due to the condition that the proportions of flows between components have to be the same for the different streams.

The difficulty with the nonconvexities noted above is that they may give rise to optimization problems involving several local optima and numerical singularities that may produce failure in the NLP algorithms. Recently there have been important efforts in the area of global optimization. Examples of algorithms are the ones proposed by McConnick (1976), Floudas and Viswewaran (1990) and Sherali and Alameddine (1992) which can be used to solve bilinear programming problems like the ones that arise in networks with multicomponent streams. For a recent review in the area of bilinear programming see Al-Khayyal (1992).

As for previous work in the design and synthesis of multicomponent process networks Mahalec and Motard (1977) and Nath (1977) developed evolutionary techniques that are based on heuristics to generate a network configuration. Floudas (1987) addressed the synthesis of separation networks with mixed products in which only sharp separators are considered. A superstructure of the process network was proposed and modelled in terms of concentrations. The resulting model is nonconvex and solved with a standard NLP algorithm with no guarantee of global optimality. Floudas and Aggaiwal (1990) solved small pooling and blending problems and sharp separation networks problems using a strategy based on Senders decomposition. In this approach only convex subproblems are solved but there is no guarantee of obtaining the global optimum. Kocis and Grossmann (1989) modelled process networks with multicomponent streams in terms of the individual component flows. They included a set of bounding constraints with binary variables to approximate the nonconvexities that are present in splitters with multiple outputs. Wehe and Westerberg (1987) studied the problem of sharp separation networks with mixed products. They proposed a search procedure that involves the

enumeration of the different separation sequences. The nonconvex equations are dropped and constraints that are valid for each particular sequence with a set of bounds over the key components are included to obtain tighter LP relaxations for each configuration. However, the number of sequences to be examined grows rapidly and there is no guarantee of global optimality*

In some particular cases the nonconvexities in the mass balances can be avoided through the introduction of binary variables. One of these cases is when single choice splitters are present in the flowsheet (Kocis and Grossmann. 1989). Here, it is possible to have a mixed integer ?fryfb: formulation for the mass balance equations in terms of the individual component flows. Another restricted case for which some nonconvexities can be reformulated is when mining within the network is only allowed for streams of the same concentration. In this form, larger network superestructures must be proposed and the concentrations of the streams are known beforehand. Integer variables are introduced to model the existence of the different streams (e.g. the mixed integer linear formulation for sharp separation networks by Floudas and Anastasiadis, 1988).

The objective of this paper is to present an efficient global optimization method that exploits the particular structure that is present in process networks with multicomponent streams (e.g. pooling and blending systems, sharp separation networks). First a relation is established between formulations based on concentrations and individual flows. This is done following the Reformulation-Linearization technique proposed by Sherali and Alameddine (1992). As will be shown, a linear relaxation Is obtained that is in the space of the concentrations and individual flows which can be used in a branch and bound search is to find the global optimum. Application to the optimal design of sharp separation systems with single feed and mixed products is considered in detail. Different preprocessing techniques that allow tightening of the relaxation problem are presented. The performance of the algorithm is reported on a total of twelve problems.

Modeling with concentrations and individual flows

Consider a process network that consists of splitters, mixers and pfrbcess units tliat are interconnected with multicomponent streams (see Fig. 1). The process iinits that arc considered in this paper are units in which the output flows of the components can be expressed as a linear relation of the inlet flows (e.g. sharp separators, reactor with^known conversion). It is possible to formulate the mathematical model of the process network in terms of the concentrations of the streams, Xj^k. Another possibility is to model the network

using flows of individual components. The former has the advantages that it provides a convenient framework for the evaluation of thermodynamic properties, and in many cases bounds can be expressed in a more natural form. A major disadvantage is that many nonconvex terms (bilinear) are involved in the mass balances for the components. Hie individual component flows formulation is often chosen since it gives rise to a larger number of linear equations and the only nonconvexities are involved in the modelling of the splitters. In these untis it is necessary to enforce that the components maintain the same concentration in each of the streams leaving the splitter. These constraints can be expressed as relations between the different components (Wehe and Westerberg. 1987). One deficiency of this representation is that since many flows can take values of zero, singularities may arise with which conventional nonlinear programming methods may have difficulties to converge. Another alternative is to introduce additional variables that represent split fractions (Kocis and Grossmann. 1989). This involves a larger number of constraints but tends to yield a formulation that numerically is better behaved.

Following are the equations that apply to the mixers, splitters and units using the two alternative representations:

<u>Mixer</u>

A mixer k consists of a set of inlet streams, M_k, and an outlet stream k (see Fig.2).

a) Concentrations

The total mass balance for a mixer k is given by:

F*= 2 F* (1)

where F* Is the total flow in stream i. The mass balance for each component j is given by the nonlinear equations,

$F^{**} \wedge \sum_{i \in M_k} Px_j^1$ forallj (2)

where x_j^1 is the concentration of component j in the stream i

b) Individual flows

Here it is only necessary to write a mass balance for each component j, given by the linear equations:

$\wedge \wedge 2_{M_k} f_j^!$ forallj (3)

where f_j^1 is the flow of component j in stream i.

<u>Splitter</u>

A splitter k has an inlet stream k and a set of outlet streams S* (see Fig. 3).

a) Concentrations

The equations for a splitter in terms of the concentrations are given by the following linear equations

$$\sum_{i \in S_k} F^i = F^k \qquad (4)$$

$$x_j^i = x_j^k \qquad \text{for all } i \in S_k \text{ and } j \qquad (5)$$

$$\sum_j x_j^k = 1 \qquad (6)$$

b) Individual flows

The mass balance for each component J is given by

$$\sum_{i \in S_k} f_j^i = f_j^k \qquad \text{for all } j \qquad (7)$$

Here, it is also necessary to enforce the condition that the streams leaving the splitter have the same proportions in flow for each component. These relations between components, which are nonlinear, can be expressed in terms of the inlet stream k and a given component J*

$$f_{j^*}^i f_j^k = f_j^i f_{j^*}^k \qquad \text{for all } i \in I_k \text{ and } j \neq j' \qquad (8)$$

A different approach consists of introducing as additional variables the split ratios ξ^i, that represent the part of the inlet flow that goes to the outlet stream i. The nonlinear equations are given by

$$f_j^i = \xi^i f_j^k \qquad \text{for all } i \in I_k \text{ and } j \qquad (9)$$

with $0 \le \xi^i \le 1$.

Process units

In this paper it will be assumed that the outlet streams, $i \in O_k$, in the process units can be expressed as linear relations of the inlet streams, $i \in I_k$ (see Fig. 4). This is for instance the case of sharp split separators, separations in which the recovery level is known, or reactors that have a fixed conversion.

a) Concentrations

The overall mass balance for process unit k is given by,

$$\sum_{i \in I_k} F^i = \sum_{i \in O_k} F^i \qquad (10)$$

The mass balance for each component j is given by the nonlinear equations:

$$\sum_{i' \in I_k} p_{ji'}^{ik} F^{i'} x_j^{i'} = F^i x_j^i \qquad \text{for all } i \in O_k \text{ and } j \qquad (11)$$

where $p_{ji'}^{ik}$ is a constant for process unit k that gives the distribution of component j in the stream $i \in O_k$ coming from streams $i' \in I_k$. For a separator unit it is required that $\sum_{i \in O_k} p_{ji'}^{ik} = 1$ and $\sum_{i' \in I_k} p_{ji'}^{ik} \le 1$. A sharp split separator is one for which $|I_k| = 1$ and $|O_k| = 2$ (top and bottom streams) and for all the components the constant $p_{ji'}^{ik}$ are either 0 or 1.

Only the **mas** balance for each component is necessary and it is given by:

$$\beta_{jr}^{ik} f_j^{r} \quad \text{forallie } O_k \text{andj} \qquad (12)$$

A model in terms of individual flows MF consists of the linear equations (3), (7) and (12) plus the nonlinear equations (8) or (9). The model in terms of the concentrations. MX includes the linear equations (1). (4), (5). (6) and (10) plus the nonlinear equations (2) and (11).

Refor"? TI totton and Linearization

In order to avoid the direct use of the nonconvex models MX and MF, there is a relation that can be established between them using the reformulation and linearization technique for bilinear programming models proposed by Shcrali and Alameddine (1992). This technique can be applied to the model MX. First, consider the bounds over the variables present in the bilinear terms (total flow, F^1 and concentrations x_j^1)

$$F^{iL} \leq F^i \leq F^{iu} \qquad (13)$$

$$x_j^{iL} \leq x_j^i \leq x_j^{iu} \qquad (14)$$

Using the bounds in (13). (14) the following constraints can be generated for the bilinear terms in (2) and (UK

$$F^i x_j^i \geq F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

$$F^i x_j^i \geq F^{iu} x_j^i + x_j^{iu} F^i - F^{iu} x_j^{iu} \qquad (16)$$

$$F^1 X_j^4 £ F^{*u} x_j^4 + x_j^L F^* - F^{*u} X_j^{IL} \qquad (17)$$

$$F^1 x_f \leq F^{iL} x_j + X_j^{4u} F^1 - F^{1L} X_j^{*u} \qquad (18)$$

In fact. McCormick (1976) has shown that the constraints in (15M 18) correspond to the convex and concave envelopes of the bilinear terms over the given bounds. The formulation is linearized by the definition of the following variables:

$$\$*¥**\{\bullet \qquad (19)$$

Tfte resultihg model which involves equations (1). (3), (4). (5), (6). (10). (12) and the constraints in (15)-{18) is a linear relaxation of the original nonconvex concentration model. MX. in which the nonlinear equations (2) and (11) have been replaced by the linear equations (3) and (12) from the individual flow model. MF. It is possible to generate additional linear constraints that are redundant to the original nonlinear model, MX, but that can be nonredundant in the linear relaxation of the model (Sherali and Alameddine, 1992: Sherali et

al.. 1992). In particular, consider equation (7) that is the linear component mass balance for the sputters in model MF. This linear equation is not present in the linear relaxation of the concentration model, MX. Take equation (4) and multiply by the valid bound constraint xf £ 0 to get

$$I_{i \in S_k} F x j \wedge F * \wedge \tag{20}$$

Using equation (5) yields.

$$t_{i \in S_k} P x j \wedge F \wedge \tag{21}$$

that can be linearized to,

$$\sum_{i \in S_k} f_j^i \; {}_{\shortparallel} f_i k. \tag{22}$$

yielding equation (7). Hence, the linear equation for the splitter is valid and it is Included. Hie nonlinear equations (8) or (9) can also be generated in a similar fashion but their linearizations are in general redundant (see Appendix A). They are only useful when the formulation df the problem provides non-trivial bounds over certain components in the outlet streams of a splitter, or when there exist some restrictions over the split ratios for the outlet streams.

Also, the constraints that relate the total flow and the individual flows of a stream can be generated for the splitters. Taking equation (6) and multiply by F^1 yields.

$$F^t \sum_j x_j^k = P \tag{23}$$

Using the constraints $xf = xj^k$ in equation (23) and linearizing with $fj^1 = F^1 xf$ yields.

$$F\text{-}I_j tf \tag{24}$$

Based on the above it is possible to obtain a reformulated model MR that involves concentrations, total flows and component flows, and which bounds the solution of the original problem. The following equations are given for model MR:

a) Objective function. 0. which is expressed in terms of individual or total flows, ..

b) Mixer equations, which are expressed ta terms of the total and individual component flows.

$$F^k = \sum_{i \in M_k} F^* \tag{1}$$

$$f_j^k = \sum_{i \in M_k} f_j^\wedge \qquad \text{for} \qquad \text{all } j \tag{3}$$

c) Splitter equations, that are expressed in terms of the individual component flows and the concentrations of the streams

$$\sum_{i \in S_k} F^i = F^k \qquad (4)$$

$$x_j^i = x_j^k \qquad \text{for all } i \in S_k \text{ and } j \qquad (5)$$

$$\sum_j x_j^k = 1 \qquad (6)$$

$$\sum_{i \in S_k} f_j^i = f_j^k \qquad \text{for all } j \qquad (7)$$

d) Process units equations, that are given in terms of the total and individual component flows

$$\sum_{i \in I_k} F^i = \sum_{i \in O_k} F^i \qquad (10)$$

$$f_j^i = \sum_{r \in I_k} \beta_{jr}^{ik} f_j^r \qquad \text{for all } i \in O_k \text{ and } j \qquad (12)$$

e) Relation between the total flow and the individual component flows

$$F^i = \sum_j f_j^i \qquad \text{for all streams} \qquad (24)$$

f) Linear estimators, relate the individual component flows with the total flow and concentrations.

$$f_j^i \ge F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

$$f_j^i \ge F^{iU} x_j^i + x_j^{iU} F^i - F^{iU} x_j^{iU} \qquad \text{for all } i \in S_k \qquad (16)$$

$$f_j^i \le F^{iU} x_j^i + x_j^{iL} F^i - F^{iU} x_j^{iL} \qquad i, k \in \text{splitters} \qquad (17)$$

$$f_j^i \le F^{iL} x_j^i + x_j^{iU} F^i - F^{iL} x_j^{iU} \qquad \text{for all } j \in J \qquad (18)$$

f) Bounds on flows and concentrations

$$F^{iL} \le F^i \le F^{iU} \qquad (13)$$

$$x_j^{iL} \le x_j^i \le x_j^{iU} \qquad (14)$$

In previous approaches (Wehe and Westerberg, 1987; Kocis and Grossmann, 1989) looser approximations of the nonlinear terms were used. In both cases, the nonconvex problem (MF) was relaxed to a linear model by dropping the nonlinear equations (8) or (9). Equations that approximate the difference relation between the components were considered (Kocis and Grossmann, 1989). They were based on the difference that exists at the inlet of the splitter between the flowrate of the components and required the introduction of binary variables.

Outline of global optimization method

Model MR can be applied to predict lower bounds to the global optimum in the optimization of pooling and blending problems and in the synthesis of separation systems. The reason is that model MR provides a valid relaxation of the original feasible region since the nonlinear equations (2) and (11) in model MX are not considered, and the valid linear equations (3), (7), (12) and (15)-(18) are included. The proposed global optimization algorithm relies on the solution of the relaxed problem MR within a spatial branch and bound enumeration. The outline of the algorithm is as follows (for a more detailed description of step 4 see Quesada and Grossmann (1993))

0. Preprocessing (optional)

Determine bounds on the variables involved in the nonconvex terms, that is total flows. F*. and concentrations, *xf*. Apply any additional preprocessing specific to the structure of the problem in order to further bound or fix variables.

1. Lower Bound

Solve model MR over a given subregion (initial subregion is the complete feasible region) minimizing a convex objective function Φ. If Φ is linear the model is an LP.

2. *Upper Bound*

Any feasible solution to the nonlinear model provides an upper bound. Heuristic techniques can be employed to obtain good feasible solutions or the original problem. MF. can be solved using the solution of model MR as a good initial point. If the solution of problem MR is feasible it provides an upper bound.

3. *Convergence*

If the lower bound of a subregion is sufficiently close or above the upper bound discard that subregion. If no subregions are left the global solution corresponds to the best upper bound.

4. *Branch and Bound*

Partition the remaining subregions into a sell of disjoint subproblems. Repeat steps 1-3 for each of the new subregions.

Remarks

The preprocessing step plays an important role in the above algorithm. It is during this step that initial bounds for the variables involved in nonconvex terms are obtained. The quality of

these bounds affects the tigthness of the lower bound since they are part of the estimator equations (15)418). Additionally, these bounds affect the performance of the algorithm because they define the search space over which the branch and bound procedure may have to be conducted.

In some cases, as described later in this paper, it is possible to exploit the particular structure of the process network and generate bounds for the variables without having |o solve any subproblems. Furthermore, during this preprocessing step additional constraints can be generated for predicting a tighter lower bound of the global optimum can be obtained.

Some of the linear mass balances and the estimator equations are redundant in the nolinear formulations, MF and MX. These equations become nonredundant in the refonnulated model, MR, and for that reason it is important to write a complete formulation of the network. However, this model can present some redundancies that can be easily identified and eliminated to reduce the size of the model. This is the case for the concentration variables used in the splitters. Model MR uses different sets of concentrations variables for the inlet and outlet variables of a split unit. In practice, it is only necessary to define the concentration of the component in the splitter and use the same variables for all the splitter streams. Also, some redundancies can occur with the total flow variables. These ones are necessary for the streams in the splitters but they might be redundant and eliminated in the other untis if they do not appear in other part of the model or in the objective function.

If the solution of model MR is feasible for the original nonconvex problem then it corresponds to the global optimal solution. When the solution to the model MR is not feasible it is necessary to follow a branch and bound procedure to search for the global optimum. This procedure requires a valid upper bound on the global optimum. This can be generated through heuristics or by solving directly the nonconvex model. For this purpose, the process network model is formulated in terms of the individual component flows and the nonconvex equations for the splitters are included. Equation (9J was also used in this work to model the splitters due to it is better numerical behavior. The solution to the model MR was used for the good initial point. In many instances, it was not possible to solve these nonlinear problems with MINOS 5.2. The nonlinear models were solved using CONOPT in GAMS 2.25.

During the branch and bound procedure a tree search is generated. Of the set of open nodes, these are the nodes that have a lower bound that is e-smaller than the current upper bound, the node with the smaller upper bound is selected to branch on. The splitter units are the units that are approximated, and of these, the splitter that has the largest difference

between its approximated and actual individual component flows is selected. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. The branching is done in the selected splitter over the concentration of the component that has the largest difference.

First, the actual concentrations for the individual component flows in the LP solution O for the splitters are calculated by,

$$z_j^k = \frac{f_j^{k*}}{F^{k*}} \quad \text{for all the inlet streams to splitter} \qquad (26)$$

the splitter unit m is then selected according to the equation.

$$m = \arg\max_{k \in split} \{ \sum_{i \in S_k} \sum_{j=1} | f_j^i - z_j^k F^i | \} \qquad (27)$$

Equation (27) represents the total difference between the LP solution for the flows after the splitter and the actual value of these flows considering the concentrations before the splitter. Once the splitter has been selected, the component in that sputter that has the largest difference. J\ is selected by.

$$j^f = \arg\max_j (\sum_{i \in S_m} | f_j^i - z_j^m F^i |) \qquad (28)$$

Hie following branching constraints are then used:

$$x_{j^f}^m \leq z_{j^f}^m \text{ and } x_{j^f}^m \geq z_{j^f}^m \qquad (29)$$

To improve the upper bound it might be necessary to solve additional nonconvex problems. These can be solved after a given number of nodes using the solution of the node with the smallest upper bound as the initial point. In this work if there was no significant change in the lower bound df the new nodes with respect to the lower bound of the parent node (< 1%) a new nonconvex problem was solved.

Example 1

Consider the following pooling and blending problem by Harveley (1978). Two streams that have components A and B are mixed in a initial mixer a then go through a splitter to obtain two streams than can be mixed with an additional stream (see Fig. 5a). Two different products can be obatined and there are constraints on the concentration of component A in these products. The objective function consists of mimizing the cost that is given by the total flow of the streams times the cost coefficients. c_t, given in Fig. 5.

This problem has two local solutions. One has an objective function Φ = -100 and consists of *otify* producing product 2. The other local solution, that corresponds to the global optimum, has an objective of 4> = -400 and here only product 1 is produced.

Model MR is formulated for this problem and the initial lower bound is Φ_L = -500. The nonlinear model, MF, is solved using the solution of model MR as the initial point and an upper bound of • a -400 is obtained* Since there is a gap between the bounds of the global solution a partition is performed- There is only one splitter that needs to be approximated and since there are only two components it is irrelevant which one is selected since the composition bounds are related (eg. x_B^L s i - x^{*u}). The actual value of the composition of A in the solution of model MR is used as the branching point (x^* = 0.0166) to generate two new subproblems. The first subproblem (x_A £ 0.0166) has a lower bound of Φ_L = -100 and the second subproblem (x^* ≤ 0.0166) has a solution of fa = -400 (see Fig. 5b). Both of these bounds are greater or equal than the upper bound, therefore the global solution has been found (x_A = 0.01).

Example 2

Tbe next example is a separation problem taken from Kocis and Grossmann (1987). The original problem has binary variables in* the formulation and they have been fixed to 1 for this example (see Fig. 6).

The top stream of the flash unit has 85% of the inlet flow of A and the bottom stream has 80% of the inlet flow of component B. In the column. 97.5% of the inlet flow of A goes to the top whereas 95% of the inlet flow of B goes to the bottom stream. The total flow to the flash unit and the column have to be greater than 2.5 and smaller than 25, whereas the total flow of each of the two feed streams has to be less than 25. The objective function is given by,

$$\Phi \text{ ss } 52 + 10 F! + 8 F_2 + F_4 + 4 F_5 - 35 P,^A - 30 P_2^B \qquad (30)$$

The initial lower bound for this problem is Φ_L = -513.22 and it is infeasible for the original NLP model. A nonconvex problem is solved using CONOPT with the solution of model MR as the initial point obtaining an upper bound of Φ = -511.87 and the relative gap is only 0.3%. Again only one splitter is present in the network and a partition can be performed using the concentration of component A in this splitter. The lower bounds for the new two subproblems are 0L = -511.87 (x_A £ 0.5121) and 0_L = -511.80 ($x_A \geq$ 0.5121). Both solutions are greater or equal than the upper bound and the global solution has been found. In the global solution F_x ss 8 and F_2 = 25, and 11% of the inlet flow to the splitter is directed to the flash. 76% to the column and the rest bypassed to P_2.

Example 3

This example corresponds to a separation problem with three feeds and three product streams. The network configuration and product specifications are given in Fig. 7. The objective function is given by

$$\phi = 4\,S_1 + 1.5\,S_2 + 4\,S_3 \qquad (31)$$

The initial lower bound is ϕ_L = 138.18 and the nonconvex problem MP is solved obtaining a solution of ϕ « 138.7. The gap between these bounds is less than 0.4 %. The global optimum for this tolerance is shown In Fig. 8.

Sharp separation networks

In order to illustrate the application of the above algorithm to a specialized case where the structure can be further (exploited, the problem that will be considered is the synthesis of separation networks with single feed and mixed products that consist of sharp separators and bypasses. It is assumed that a single feed with N components must be separated into K specified multicomponent product streams. The components are ordered from the lightest to the heaviest.

A modification of the superstructure proposed by Floudas (1987) for this problem is used (see Fig. (9)). The superstructure consist of N-l separators. Separator 1 performs the task of removing component number 1 to number 1 at the top of the separator and components number i+1 to N at the bottom of the separator. The feed to the network is split into N-l streams, F_t. that go to the separators and K streams, a_k. that bypass the network to go to the products. Each stream F< is mixed before the separator i with streams that come from the top and bottom streams from the other separators to obtain the inlet stream to separator i, St.

The outlet streams of separator i are the top. T_{l9} and the bottom, B_t. These streams, T« and B{, are each split into streams, PT_t^k and PB<^{k} respectively, that go to the K products and into streams, KIV' and RB/\ that are redirected to the other separators. The top stream of separator 1, T_t, can be redirected only to the separators 1 to i-1 since it can only contain components number 1 to i. It would not be optimal to send part of this stream to any separator from i+1 to N since no separation would be achieved and a bypass of these separators would achieve the same separation with smaller (lows. *KTf* is the flow redirected from the top of separator i to separator i\ In the same fashion that with the top stream, the bottom of separator i, B_t, can be redirected only to separators i+1 to N-l since it can only

contain components that are separated by these sharp separators. $RB_i^{i'}$ is the redirected flow from the bottom stream of separator i to separator i'.

Model

It will be assumed that the objective function can be expressed as a linear function that depends on the total flow to each separator. The model expressed in terms of concentrations and total flows has the following form:

$$\min \Phi = \sum_{i=1}^{N-1} c_i S_i \qquad (32.1)$$

$$\text{s.t.} \quad Feed = \sum_{i=1}^{N-1} F_i + \sum_{k=1}^{K} \alpha_k \qquad (32.2)$$

$$f_{ij} = F_i z_j \quad \text{for all i and j} \qquad (32.3)$$

$$S_i = F_i + \sum_{i'=1}^{i-1} RB_{i'}^{i} + \sum_{i'=i+1}^{N-1} RT_{i'}^{i} \quad \text{for all i} \qquad (32.4)$$

$$S_i xs_{ij} = f_{ij} + \sum_{i'=1}^{i-1} RB_{i'}^{i} xb_{i'j} + \sum_{i'=i+1}^{N-1} RT_{i'}^{i} xt_{i'j} \quad \text{for all i and j} \qquad (32.5)$$

$$S_i = T_i + B_i \quad \text{for all i} \qquad (32.6)$$

$$T_i xt_{ij} = S_i xs_{ij} \quad \text{for all i and ord(j)} \le i \qquad (32.7)$$

$$xt_{ij} = 0 \quad \text{for all i and ord(j)} > i \qquad (32.8)$$

$$B_i xb_{ij} = S_i xs_{ij} \quad \text{for all i and ord(j)} > i \qquad (32.9)$$

$$xb_{ij} = 0 \quad \text{for all i and ord(j)} \le i \qquad (32.10)$$

$$T_i = \sum_{i'=1}^{i-1} RT_i^{i'} + \sum_{k=1}^{K} PT_i^k \quad \text{for all i} \qquad (32.11)$$

$$B_i = \sum_{i'=i+1}^{N-1} RB_i^{i'} + \sum_{k=1}^{K} PB_i^k \quad \text{for all i} \qquad (32.12)$$

$$P^k = \sum_{i=1}^{N-1} PT_i^k + \sum_{i=1}^{N-1} PB_i^k + \alpha_k \quad \text{for all k} \qquad (32.13)$$

$$p_{kj} = \sum_{i=1}^{N-1} PT_i^k xt_{ij} + \sum_{i=1}^{N-1} PB_i^k xb_{ij} + \alpha_k z_j \quad \text{for all k and j} \qquad (32.14)$$

$$\sum_{ord(j)=1}^{i} xt_{ij} = 1 \quad \text{for all i} \qquad (32.15)$$

$$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1 \quad \text{for all i} \qquad (32.16)$$

$$\sum_{ord(j)=1}^{N} xs_{ij} = 1 \quad \text{for all i} \qquad (32.17)$$

$$S_i, T_i, B_i, F_i, RT_i^{i'}, RB_i^{i'}, PT_i^k, PB_i^k, \alpha_k, f_{ij}, xs_{ij}, xt_{ij}, xb_{ij} \ge 0$$

The parameters Feed, z_j, P^k and p_{kj} represent the total feed, composition of the feed, total flow of product k and component flow of component j in product k, respectively. The variables xs_{ij}, xt_{ij} and xb_{ij} are the concentrations of component j in the inlet stream to separator i, top of separator i and bottom of separator i, respectively.

The objective function (32.1) is a linear function of the inlet flow to the separators. Equation (32.2) is the total mass balance in the inlet splitter and equation (32.3) is the component mass balance. Equations (32.4) and (32.5) are the total and component mass balances for the mixer i before the separator i. The material balances for separator i are given by equations (32.6), that is the total mass balance for the separator, equations (32.7) and (32.8) that are the component balances for the top stream and indicate that nothing from components number i+1 to N-l is in the top of the separator, and equations (32.9) and (32.10) that are the component mass balances for the bottom streams. Equations (32.11) and (32.12) are the overall mass balances for the splitters of the top and bottom streams after separator i. The equations that state that the concentrations of the outlet streams should be the same that the inlet stream in a splitter have been already substituted. Finally, equations (32.13) and (32.14) are the overall and component mass balances for the mixer for product k.

Model (32) corresponds to a formulation of the type of model MX where the distribution coefficients are known and restricted to 0 or 1. Some simplifications have been made to avoid including many irrelevant variables (e.g. not to define concentrations for the streams that go the top i to product k). Although, some of the linear constraints in this formulation are redundant, they can become nonredundant in the linear relaxation as will be shown in Example 4.

Equations (32.5), (32.7), (32.9) and (32.14) involve nonconvex terms. This model can be reformulated as in model MR by introducing individual component flows and the linear equations (15)-(18) and (7) according to the approach illustrated eaiiier to obtain a model in the form of model MR. The resulting reformulated model is as follows,

$$\min \ \Phi' = \sum_{i=1}^{N-1} c_i S_i \qquad (33.1)$$

$$\text{st} \quad \text{Feed} = \sum_{i=1}^{N-1} F_i + \sum^{K} \alpha_k \qquad (33.2)$$

$$f_{ij} = F_i z_j \qquad \text{for all i and j} \qquad (33.3)$$

$$S_i = F_i + \sum_{i'=1}^{i-1} RB_{i'}{}^{i} + \sum_{i'=i+1}^{N-1} RT_{i'}{}^{i} \qquad \text{for all i} \qquad (33.4)$$

$$s_{ij} = f_{ij} + \sum_{i'=1}^{i-1} rb_{i'j}{}^{i} + \sum_{i'=i+1}^{N-1} rt_{i'j}{}^{i} \qquad \text{for all i and j} \qquad (33.5)$$

$$S_i = T_i + B_i \qquad \text{for all i} \qquad (33.6)$$

$$t_{ij} = s_{ij} \qquad \text{for all i and ord(j) £ i} \qquad (33.7)$$

$$t_{ij} = 0 \qquad \text{for all 1 and ord(j) > 1} \qquad (33.8)$$

$$b_{ij} = s_{ij} \qquad \text{for all i and ord(j) > i} \qquad (33.9)$$

$$t_{ij} = 0 \qquad \text{for all i and ord(j)} \leq \text{i} \qquad (33.10)$$

$$T_i = \sum_{i'=1}^{i-1} RT_i^{i'} + \sum_{k=1}^{K} PT_i^{k} \qquad \text{for all i} \qquad (33.11)$$

$$B_i = \sum_{i'=i+1}^{N-I} RB_i^{i'} + \chi PB_i^{k} \qquad \text{for all 1} \qquad (33.12)$$

$$P^k = {}^{N-I} PT_i^{k} + \sum_{i=1}^{N-1} PB_i^{k} + \alpha_k \qquad \text{for all k} \qquad (33.13)$$

$$p_{kj} = \sum_{i=1}^{rt-i} pt_{ij}^{k} + \sum_{i=1}^{N-1} pb_{ij}^{k} + \alpha_k z_j \qquad \text{for all k and J} \qquad (33.14)$$

$$t_{ij} = \sum_{i'=1}^{i-1} rt_{ij}^{i'} + \sum_{k=1}^{K} pt_{ij}^{k} \qquad \text{for alii and j} \qquad (33.15)$$

$$b_{ij} = \sum_{i'=i+1}^{N-I} rb_{ij}^{i'} + \sum_{k=1}^{K} pb_{ij}^{k} \qquad \text{for all i and j} \qquad (33.16)$$

$$\sum_{ord(j)=1}^{i} xt_{ij} = 1 \qquad \text{for all i} \qquad (33.17)$$

$$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1 \qquad \text{for all i} \qquad (33.18)$$

$$T_i = \sum_{ord(j)=1}^{i} t_{ij} \qquad \text{for all i} \qquad (33.19)$$

$$B_i = \sum_{ord(j)=i+1}^{N} b_{ij} \qquad \text{for alii} \qquad (33.20)$$

$$PT_i^{k} = \sum_{ord(j)=1}^{i} pt_{ij}^{k} \qquad \text{for all i and k} \qquad (33.21)$$

$$PB_i^{k} = \sum_{ord(j)=i+1}^{N} pb_{ij}^{k} \qquad \text{for all 1 and k} \qquad (33.22)$$

$$\text{" " •-S/} \qquad \text{for all i and i' < i} \qquad (33.23)$$

$$RB_i^{i'} = \sum_{ord(j)=i+1}^{N} rb_{ij}^{i'} \qquad \text{for all i and i' > i} \qquad (33.24)$$

Equations (15-18) forty. rt_{rj}^{1} and pt_{y}^{k} in terms of xtg and the total flow of its respective stream.

Equations (15-18) for by. rbq^{1} and pb_{tl}^{k} in terms of xb_{ij} and the total flow of its respective stream.

S,.T,, B,. F,. RT,'. RB,^{f}. PT,^{k}. PB,^{k}, Ok. f_{tt}. xs,j. xU_t. $xb_{(J}$ SO

Sg. t,j. b,j pti,^{k}. pb„^{11}. rt,,$^{1'}$. rb„$^{1'}$ ≥ 0

It is not necessary to Include equations (15)-(18) for the inlet component flows to the separator, Sq, since the variables xs,j only appear in these equations. Also, the component flows, s,j, only appear in mixers and sharp separators units which can be exactly represented in terms of the individual component flow equations (33.5). (33.7) and (33.9). Equations (33.15)-(33.16) that are the component mass balances for the splitters of top and bottom streams have been included accordingly to the reformulation previously presented. Equations (33.19M33.24) relate the total flow and the individual component flows for the splitter streams.

Preprocessing

Hie proposed superstructure (Fig. 9) allows to bypass certain amount of the feed to the product k, a*, without having to go through the separation network. The amount of the product k that is not bypassed has to be processed in the separation network and it will be denoted as the 'residual product*. Hence, the total 'residual product k' is given by (P^k - a_k) and has the component flows given by (p^ - o^z,)(see Fig. 10).

The global optimal solution of model (32) is a network in which all the 'residual products'.have at least one component with a zero flow. The reason that it is not optimal to separate a stream in the network and later to remix it. The same degree of separation can be achieved using a bypass that does not incur any cost in the objective function.

Consider the second separator in the solution obtained by Floudas (1987) to his second example (see Fig. 11). For this subnetwork of the complete structure the upper 'residual product* has components B and C present. The components are being separated and remixed again. The same outlet flows can be obtained with a smaller input flow to the separator as it is shown in Fig. 11. Note that both 'residual products' have components with zero flow.

It should be clear that if there was not a component with zero flow in the 'residual product[1], then there is part of this stream that could have been obtained by just bypassing the network. This in turn does not incur in any cost, whereas going through the network has a positive cost. The above condition gives a lower bound for the bypass to each product. This also corresponds to the largest amount that can be bypassed since all the flows in the 'residual flow* have to be positive. In this form the bypass can be precalculated without affecting the global optimality of the solution.

The bypass to product k is given by the maximum amount that can be sent to product k without having a negative flow; that is.

a„ = min, [^4 (34)

where Zjis the feed composition and p_{kj} is the flow of component j in product k. The component flows for 'residual product' k. Tkj are given by.

$$\gamma_{kj} = p_{kj} - \alpha_k z_j \qquad (35)$$

Key component bounds

Wehe and Westerberg (1987) proposed using lower bounds for the (low of the key components in separator L These bounds are based on the fact that separator i is the only unit that can perform the task of separating component number 1 from component nutaber 1+1. They are redundant for the nonlinear formulation (32) but they are relevant for the linear relaxation in (33). To calculate them, it is necessary to determine in each product what is the difference between the two key components of separator i with respect to the concentrations in the feed. The lower bounds in separator i' for the flow of the key components in the top (component Jl) and bottom (component J2) streams are given by:

$$t_{ij1} \geq \sum_{k=1}^{K} \{ \gamma_{kj1} - z_{j1} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for alii} \qquad (36)$$

$$b_{ij2} \geq \sum_{k=1}^{K} \{ \gamma_{kj2} - z_{j2} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{foralli} \qquad (37)$$

where Ykj is given by (35). It is important to include both bounds in the relax model (33) since there is no guarantee that the inlet flow to separator i has the same proportion between the key components as the feed. It is not known in which part of the sequence separator i will be placed, and it can be after a splitter that is not being approximated correctly.

The bounds in (36) and (37) can be extended to separation of components that are not adjacent in the feed. Consider component number i and component number i+3. There are three separators that can perform this task/separators i. i+1 and i+2. Cuts of the following form can be obtained.

$$t_t\, i + t_M\, i + t|{*}a\ 12 \sum^{K} \{.Dd - zt \min_{ord(j),< or H3} [\frac{\gamma_{ki}}{r}]\} \qquad \text{for all i} \qquad (38)$$

$$b_{i1} + b_{i+1}\, i + b_{M\,i}\ 2 \sum^{K} \{Tki{*}3 - z,{*}_3 \text{ minors}_{OM}{*}3\ \frac{y_{kl}}{i})) \qquad \text{for all i} \qquad (39)$$

Equations as the ones in (38) and (39) can be redundant compared to equations (36) and (37) and it is possible to detect this before solving the problem.

Relativeflowrate constraints

These constraints are used when the relation between the flowrates of two components is known. In particular, consider component A in the la:st column of the network (see Fig. 12). None of the redirected streams contains component A. Therefore, the relative flowrate of component A with respect to the other components in the top stream has to be smaller than in the feed. This relation should remain valid after the top stream is split to the products and redirected flows.

Ill the separator previous to the last one, N-2, all the streams do not have component A except the one coming from the top of the last column. For this one it is already known that the relative flow of component A with respect to the other components is smaller than in the feed. This type of analysis can be done for component A and component N in all the columns yielding th$ following linear constraints for the splitters.

for all i and k

$$z_A\, pt_{ij}^k - z_j\, pt_{iA}^k \ge 0 \qquad \text{for } j \ne A \text{ and } ord(j) \le i \qquad (40)$$

$$z_N\, pb_{ij}^k - z_j\, pt_{iN}^k \ge 0 \qquad \text{for } ord(j) \ne N \text{ and } ord(j) > i \qquad (41)$$

for all i and l' > i

$$z_A\, rt_{ij}^{l'} - z_j\, rt_{iA}^{l'} \ge 0 \qquad \text{for } j \ne A \text{ and } ord(j) \le i \qquad (42)$$

for all i and l' < i

$$z_N\, rb_{ij}^{l'} - z_j\, rb_{iN}^{l'} \ge 0 \qquad \text{for } ord(j) \ne N \text{ and } ord(j) > i \qquad (43)$$

Bounds on concentrations and total Jlows

The approximations (15-18) require bounds for the total flows and component concentrations In the splitters. The lower bound for the total flow of the top and bottom streams is given by the lower bound of the key components obtained in equations (36) and (37). For the outlet streams of the splittters, that are the redirected streams and the streams that go to the products, the lower bound is zero. The upper bound for the total flow of the top and bottom streams is given by the feed to the network of the components that can be present in each stream .that is.

$$T_i^U = \sum_{ord(j)=1}^{i} Feed \sum_k \alpha_{kj} z_k, \qquad \text{for all } i \qquad (44)$$

$$B_i^U = \sum_{ord(j)=i+1}^{N} Feed \sum_k \alpha_{kj} z_k \qquad \text{for all } i \qquad (45)$$

The upper bound for the streams after the splitter are given by.

$RT_i^{i'u} = T_i^u$ for all i and $i' < i$ (46)

$RB_i^{i'u} = B_i^u$ for alii aridi'> i (47)

$PT_i^k = \sum_{and(j)=1}^{i} Y_{kj}$ for all i and k (48)

$PB_i^k =$ Jb? for all i and k (49)

The lower bounds for the concentrations are zero except for fee key components in the separator for which the lower bounds are given by the lower bound of its flow divided by the upper bound of the total flow of that stream. The upper bounds in the concentrations are given by one minus the lower bounds of the other components.

The solution of the linear programming model (33) provides a lower bound to the global optimum since this model is a valid relaxation of the nonconvex model (32). This lower bound is obtained by solving the LP model for the 'residual products' in (35) with the addition of the valid constraints (36)-(43).

The upper bounds are generated solving model (32) in terms of the individual flows for the 'residual products'. When additional nonconvex problems are solved to improve the current upper bound it can happen that very similar initial point are generated. In this case, a new nonconvex NLP is solved in which bounds over the total inlet flows to the separators (S,) are included. For this purpose the values of these variables in the LP solution (S,*) arc used such that the current incumbent solution is no longer feasible.

Example4

Consider the 3 component example proposed by Floudas and Aggarwal (1990). An equimolar feed has to be separated into two products as show in Fig. 13. The objective function is given by

$$\phi = 0.2395 + 0.00432\, S_1 + 0.7584 + 0.01517\, S_2 \qquad (50)$$

The bypass to products 1 and 2 can be calculated according to equation (34) and the 'residual product' component flows are obtained through equation (35) (see Fig. 14). The total bypass to product 1 is $\alpha_1 = 90$ and the bypass to product 2 is $\alpha_2 = 150$ and the feed has a concentration of $z_A = 1/3$, $z_B = 1/3$ and $z_C = 1/3$. In this form the 'residual product' 1 is $Y_{1A} = 0$, $Y_{1B} = 20$ and $Y_{1C} = 0$ and the 'residual product' 2 is $Y_{2A} = 20$, $Y_{2B} = 0$ and $Y_{2C} = 20$. Additionally, lower bounds on the flow of the key components in both separators are obtained

using equations (35M36). The key components in separator 1 are component A in the top and its flow has to be at least 20 and component B in the bottom has to have at least a flow of 20. In the top stream of the second separator at least 20 units of component B have to be separated from 20 units of component C in the bottom stream. It is important to note that after preprocessing the network several suboptimal solutions have been cut off. One of these suboptimal solutions for this particular data is a parallel configuration of both separators (there are situations in which a parallel configuration can correspond to the global solution as will be shown in example 5). In this example the direct or indirect sequence have a lower objective function. Both of these configurations are local solutions with an objective function value of 4 = 1.8639 for the direct sequence and $ = 2.081 for the indirect one. In some instances, MINOS 5.2 had problems converging even in this small example.

The LP (33) is formulated for this problem, giving a lower bound of ^ = 1.8639. The approximations are exact and therefore this solution is a feasible solution of model (32) proving that it corresponds to the global optimum. Hence, convergence is achieved in one iteration. The optimum solution corresponds to the direct sequence shown in Fig. 15. It should be noted that if the linear mass balances for the mixer for product 2 were not considered since they are redundant for the nonlinear formulation, a lower bound in the relaxed model of $\$_L$ = 1.12 is obtained. This shows that It is relevant to include all the mass balances in the linear model in order to tighten the lower bound.

ExompteS

In the approach proposed by Wehe and Westerberg (1987) for the case of 3 components only the direct and indirect sequences are considered and both options can be modelled as LP problems since no mixing is required for these separation networks. However, this example shows that parallel configurations can be also globally optimal and that they are not excluded by the method proposed in this paper. To be able to consider parallel configurations or any combination of parallel with direct or indirect sequences it is necessary to model a superstructure in which mixing is allowed (like in the structure used in Fig. 13). Here, nonconvexities arise in the mass balance equations after the separators.

Consider that an equimolar feed is to be separated into the two different products given in Fig. 16. The objective function is to minimize the sum of the total flows into the separators. The same procedure that in the previous example is followed and the bypass can be precalculated by equation (34). The solution to the model (32) yields Φ_L = 12 and since it is a feasible solution of model (32) it corresponds to the global optimum (see Fig. 16). Note that the solutions for the direct or indirect sequences have an objective function of $ = 16.

Branch and Bound

If there is a gap between the lower ai}d upper bound, a branch and bound search is performed. It is only necessary to do the search over the variables involved in the nonconvex terms. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. In this way. it is necessary to check the approximation for the concentrations in the splitters of the top and bottom strear^s of the separator. Equations (26)-(29) for the splitters of top and bottom streams are used to perfom the branch and bound search.

Results

Table 1 summarizes the results of the earlier examples 1 to 3 and of the sharp separation network examples 4 to 12. The number of variables is the total number of variables that are included in the reformulated and relaxed model (33) for that example. The lower bound is the initial bound that is obtained by solving model (33) over the entire feasible space. The inital gap represents the percentage difference between the initial lower and upper bounds. When there is a zero initial gap it means that the first relaxed solution is feasible in the original problem thereby corresponding to the global solution. The column for nodes gives the total number of subproblems that where solved before converging to the global solution. A relative tolerance of 0.01 was used, except for example 2 were exact convergence was obtained after branching and example 12 for which a tolerance of 0.02 was used. It is important to note that the initial lower bound is tight and that it corresponds to a good estimation of the global solution. The largest differences are for example 1 with a 25% of difference and for example 12 with a 7% difference. The LP time refers to the time used to solve each relaxed model and the NLP time is the time used for solving a nonconvex model. It is possible to do updates using the previous LP solution and in this form have a more efficient implementation. The times are in seconds and the problems were solved on an IBM RS600/530 using GAMS 2.25 (Brooke et al. (1988)). MINOS 5.2 was used to solve the LP problems and CONOPT for the nonconvex NLP problems. A brief description of the example problems 6 to 12 is given below. It includes the specific data for the problem, the objective function and the topology of the network that is the global solution.

Example 6

This example corresponds to example 2 from Floudas (1987). In this case a linear objective function with the same cost coefficients is used and it is given by,

$$(M2.5S_1+3.0S_2+1.5S_3 \qquad (51)$$

The data for the composition of the products is given in Table 2.

The initial lower bound is $\phi_L = 54.25$ and an upper bound of $\phi = 55.5$ is obtained by solving the nonconvex problem. A partition of the feasible region is performed using the composition of component D in the bottom stream of separator 1. The first subproblem ($x \geq 0.166$) yields a lower bound of $\phi = 55.45$ and the second subproblem ($x \leq 0.166$) has a solution of $\phi_L = 55.8$. The latter is greater than the upper bound and the former is less than 1% of the global solution (see Fig. 17).

Example 7

This example is taken from Floudas (1987). The data for this problem is given in Table 3 and the linear objective function is given by:

$$\phi = 2.5\, S_1 + 3.0\, S_2 + 1.2\, S_3 \tag{52}$$

The initial lower bound is $\phi_L = 32.7$ and it provides a feasible solution to the nonconvex problem. In this form the global solution (see Fig. 18) is obtained in one iteration. It is interesting to see that this solution also provides a better objective function for the concave objective function used by Floudas (1987) ($\phi = 10.65$ versus $\phi = 13.68$ which is 28% higher)

Example 8

This four component problem is taken from Wehe and Westerberg (1987). The data for the products is given in Table 4 and the objective function has the following form:

$$\phi = 5.0 + 0.5\, S_1 + 4.0 + 0.3\, S_2 + 6.0 + 0.7\, S_3 \tag{53}$$

The first relaxed subproblem has a solution of $\phi_L = 26.76$ and it is infeasible for the nonconvex problem. A nonconvex problem is solved using CONOPT with the LP solution as the initial point. An upper bound of $\phi = 26.79$ is obtained corresponding to the global solution (see Fig. 19) within a 0.1%.

Example 9

This example corresponds to example 1 from Wehe and Westerberg (1987). Table 5 provides the data for the product flows and the objective function is given by:

$$\phi = 5.0 + 0.5\, S_1 + 9.0 + 1.0\, S_2 + 3.0 + 0.4\, S_3 + 6.0 + 0.6\, S_4 \tag{54}$$

A initial lower bound of $\phi_L = 85.16$ is obtained and the upper bound is $\phi = 85.65$. The difference is 0.5% and the global solution (see Fig. 20) is obtained in one iteration.

Example 10

This problem is taken from Flotidas (1987) and the data is given in Table 6. The objective function is given by,

$$\Phi = 1.2\, S_1 + 3.0\, S_2 + 2.5\, S_3 + 1.5\, S_4 \qquad (55)$$

The initial lower bound is Φ_L = 156.56 and the upper bound is Φ_U = 179.08. After 5 nodes the global solution of Φ = 159.48 (see Fig. 21) is obtained.

Example 11

The data for this 6 component problem are given in Table 7 and the objective function has the following form:

$$\Phi = 1.5\, S_1 + 3.0\, S_2 + 2.0\, S_3 + 1.0\, S_4 + 4.0\, S_5 \qquad (56)$$

The initial lower bound is Φ_L = 173 and the upper bound is Φ = 179.11. After five nodes the global solution is obtained (see Fig. 22).

Example 12

This is a 6 component 4 products problem and the data are given in Table 8. The objective function is:

$$\Phi = 5.0\, S_1 + 3.0\, S_2 + 2.0\, S_3 + 2.5\, S_4 + 4.0\, S_5 \qquad (57)$$

The initial lower bound is Φ_L = 362 and the initial upper bound is Φ_U = 415.6. The global solution of Φ = 388 (with a 2% optimality gap) is obtained after 33 nodes (see Fig. 23).

Conclusions

A general procedure for the global optimization of process networks with multicomponent streams has been proposed. The basic idea relies on a relaxed LP model that is obtained through reformulation-linearization techniques that establish a clear relation between the component flow and the composition models for mass balances. The reformulated model combines both of these providing tighter lower bounds than other relaxations proposed in the previous work. The relaxed linear model has been embedded in a branch and bound procedure to obtain the global optimal solution.

As has also been shown, the algorithm can be further specialized to take advantage of the particular structure of sharp separation networks with single feed and mixed products. Here, it is possible to preprocess the problem to reduce the space over which the search is

conducted. The bounds that are necessary for the estimator functions in the relaxed model can be obtained without having to solve any subproblems. Different types of linear approximations that are nonredundant to the relaxed model are included to obtain a tighter lower bound.

Twelve examples for both general process networks and for sharp separation networks have been presented to illustrate the performance of the algorithm. As has been shown, only a small number of nodes are commonly needed in the branch and bound search to identify the global or e-global solution. Moreover, in many cases the initial lower bound is either the exact solution or a very good approximation to the global solution.

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Appendix A. Reformulation-Linearization to obtain the nonlinear constraints in **model MF**

The nonlinear equations in **model** MF, that can be expressed either as (8) or (9), can also be generated from model MX. For this purpose take the concentration model MX and consider equation (5),

... $x_j^k = x_j^l$ (A.1)

multiply by the valid bound constraint Xj-^{k} £ 0

xfxfmxfzp **(A.2)**

Use equation (5) for **component** j\

x,^{k}Vi^l*f^k** 1A-3

Multiply by **the valid bound** constraints $F^k \geq 0$ and $F^l \geq 0$,

F^JP^-F^F*^ tA.41

that it is linearized to yield.

fffJJsfj'ff (A.5)

which is precisely equation (8) for the splitters in the individual flow model MF.

Consider again equation (5).

$x_j^k = x_j^l$ (A.6)

multiply by the valid bound constraints $F^k \geq 0$ and $F^* \geq 0$,

$F^k x_j^k F^l = F^l x_j^l F^k$ (A.7)

that can be linearized to yield,

ffF^f/F* (A.8)

Define the split fraction ? to be,

?=|* (A.9)

Equation (A.8) can then be expressed as

f/ = ? fj^k (A.10)

which corresponds to equation (9).

Hence, the nonlinear equations (8) and (9) are redundant to model MX. Their linear approximations in general are also redundant in the linear reformulated model MR Consider equation (A.10), similarly to (15) one of its linear approximations is given by.

$f_j^l \geq \xi^{lL} f_j^k + \xi^l f_j^{kL} - \xi^{lL} f_j^{kL}$ (A.11)

If there are no particular restrictions in the splitters, then the bounds for the split fraction variable are $0 \leq \xi \leq 1$ and using them in (A. 11) yields.

$$f_j^l \geq f_j^{kL} \xi^l \qquad \text{(A.12)}$$

The bound for the individual component flow is given by $f_j^{kL} = x_f^{L} F^{kL}$: also $x,^k = x,^4$ and $£^f$ = $\underline{F}^1$
pt, which leads to.

$$f_j^l \geq x_j^{lL} F^l \frac{F^{kL}}{F^k} \qquad \text{(A.13)}$$

The estimator (15) for the same conditions ($F^{*L} = 0$) is given by

$$f_j^l \geq x_j^{lL} F^l \qquad \text{(A.14)}$$

Since the factor -^*r* is always less or equal than 1, equation (A. 13) is redundant. A similar analisis can be performed for the other estimators. Only when more specific bounds over the split fractions or the individual component flows are known, will these additional estimators be non redundant.

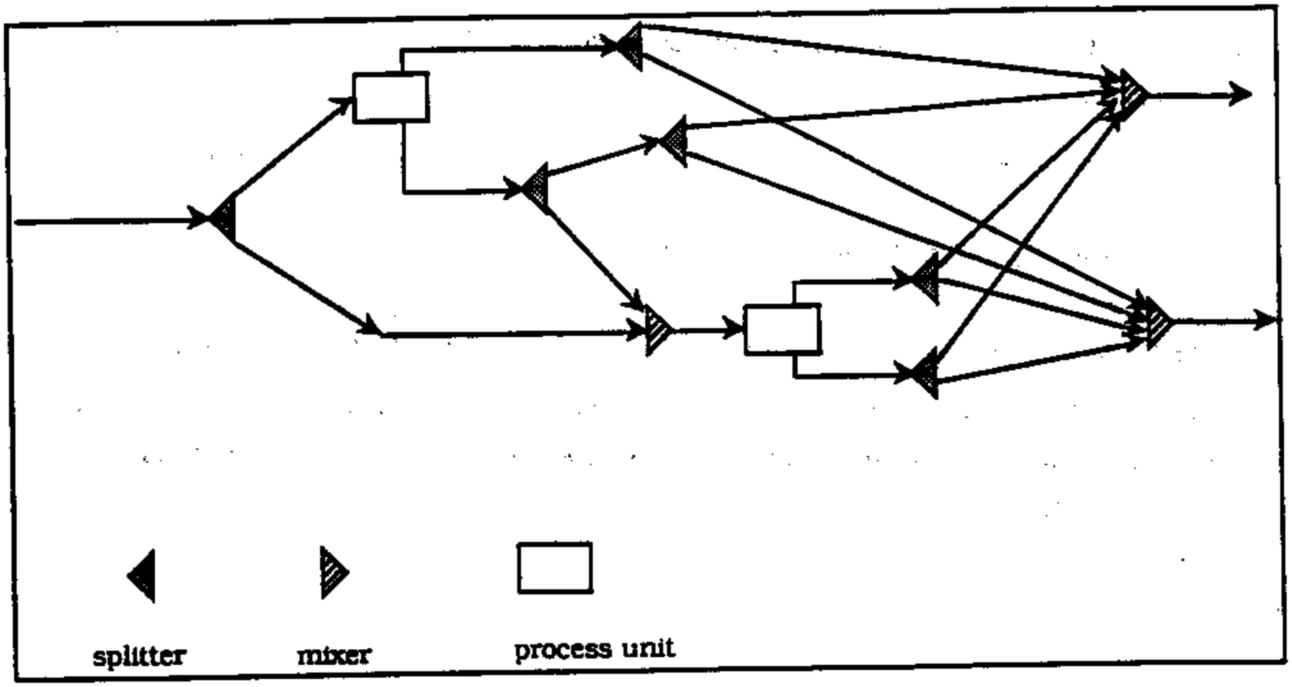


Figure 1. Process network with units, splitters and mixers.

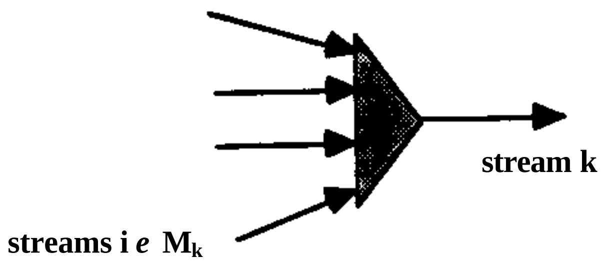


Figure 2. Mixer module.

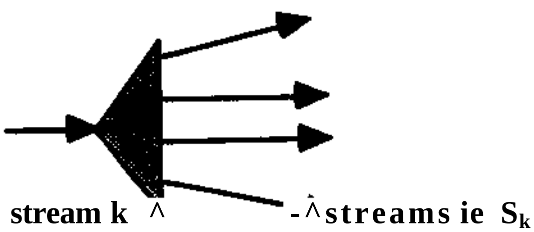


Figure 3. Splitter module.

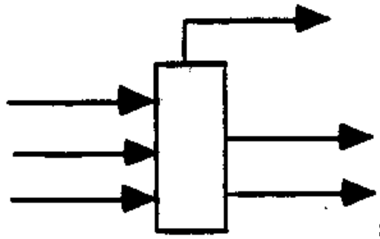
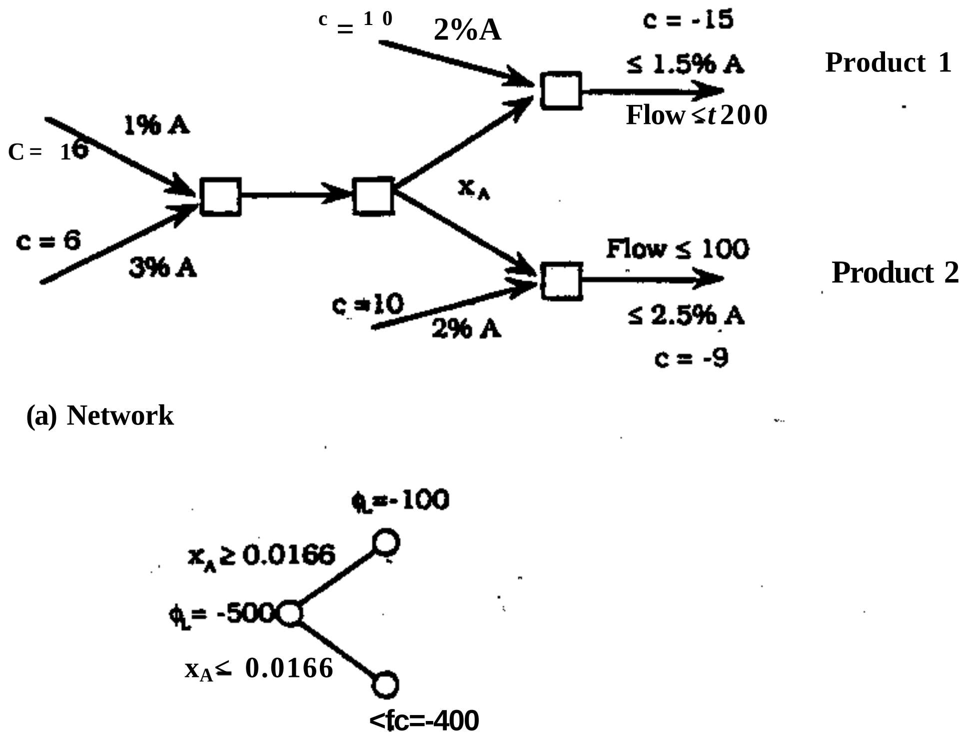


Figure 4. Process unit module.



(b) Tree search

Figure 5. Network and branch and bound search for example 1

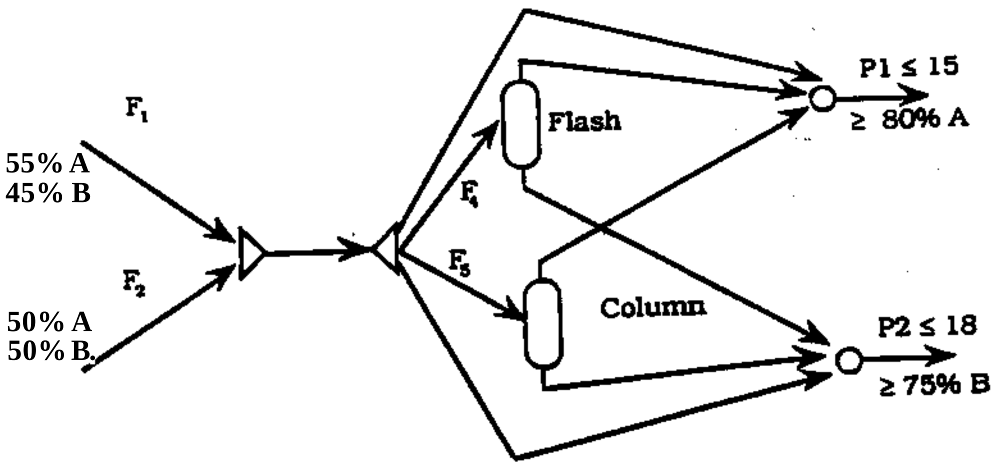


Figure 6. Network for example 2.

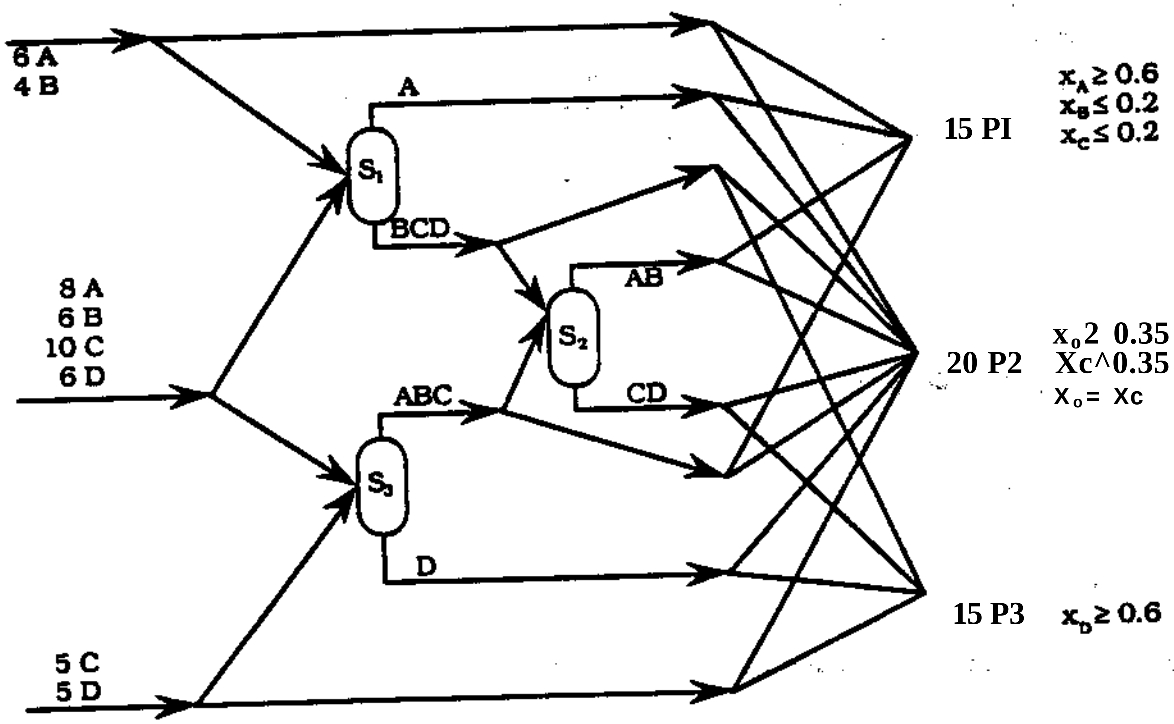


Figure 7. Network for example 3.

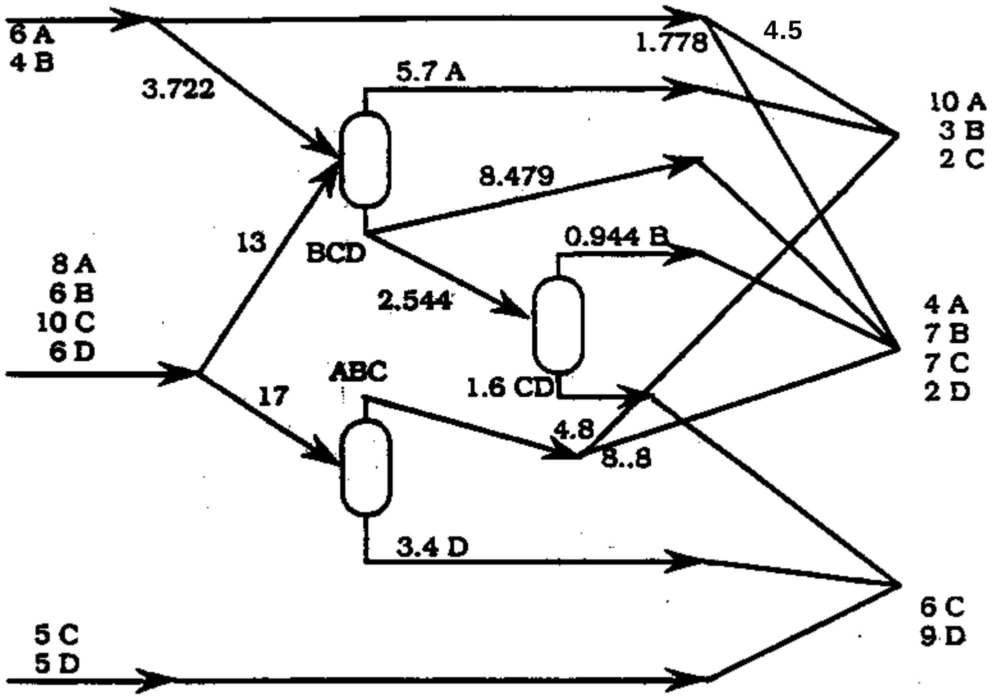


Figure 8. Optimal network for example 3.

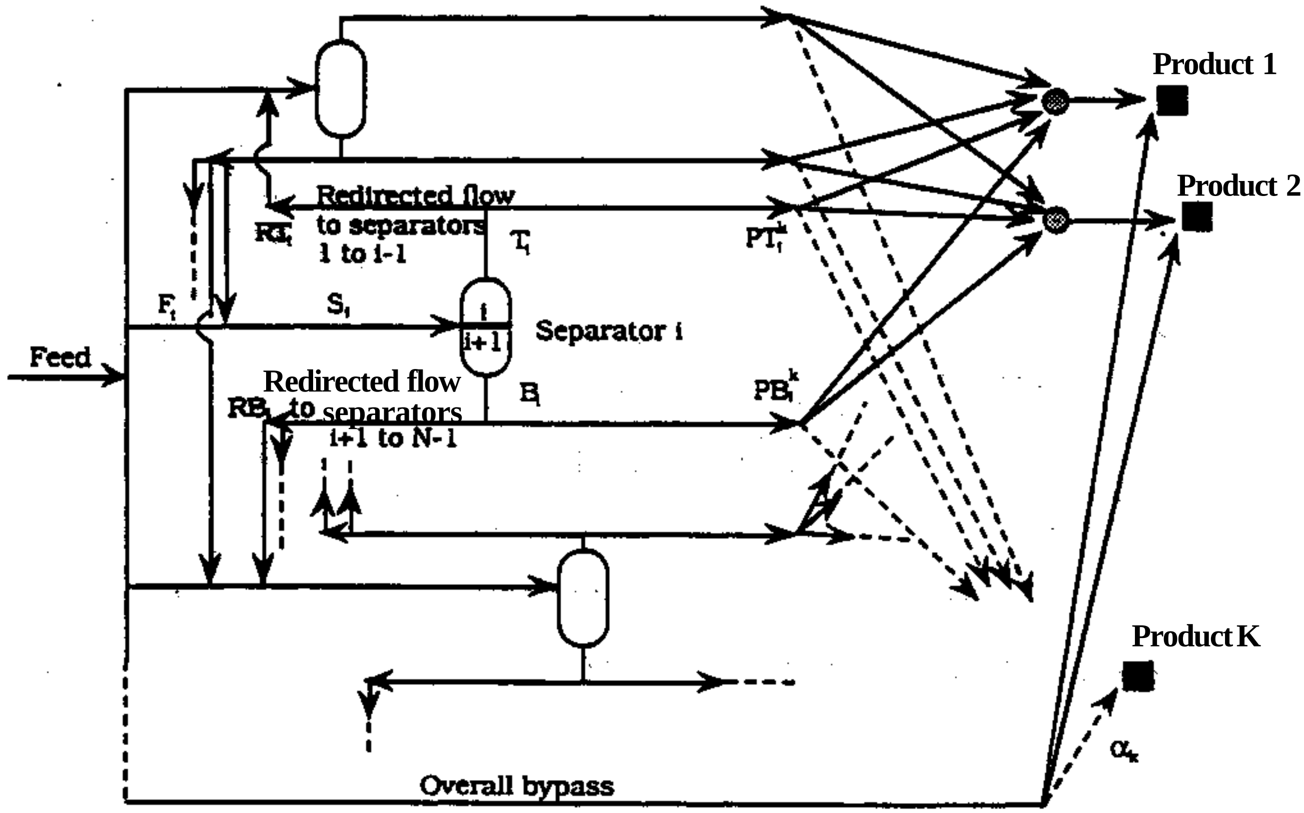


Figure 9. Superstructure for separation with sharp splits and mixed products.

Global Optimization of Process Networks with Muiticomponent Flows

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EDRC 06-164-94

Global Optimization of Process Networks with Multicomponent Flows

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Abstract

This paper deals with the global optimization of networks consisting of splitters, mixers and linear process units and which involve multicomponent streams. Examples include pooling and blending systems and sharp separation networks. A reformulation-linearization technique is first applied to concentration and flow based models in order to obtain a relaxed LP formulation that provides a valid lower bound to the global optimum. This formulation is then used within a spatial branch and bound search. The application of this method is considered in detail for sharp separation systems with single feed and mixed products. Numerical results are presented on twdve test problems to show that only few nodes are commonly required in the branch and bound search.

Introduction

A common source of noneonvexities in the synthesis and design of processes, as well as in flowsheet optimization, are the material flow equations for multiccmponent streams. These nonconvex equations involve bilinear terms and they arise in the mass balance equations when the compositions are unknown. There are different equivalent formulations for this type of networks. One alternative is to formulate the mass balance equations in terms of component concentrations. In this form bilinear terms are present in the equation* for the mixer units and the different process units (e.g. sharp separators). A second alternative is to express the mass balances in terms of flows of individual components. This option has the advantage that it involves a ^*naUmr* number of nonlinear equations. However, the modelling of the splitter units involves bilinear terms that arise due to the condition that the proportions of flows between components have to be the same for the different streams.

The difficulty with the nonconvexities noted above is that they may give rise to optimization problems Involving several local optima and numerical singularities that may produce failure in the NLP algorithms. Recently there have been important efforts in the area of global optimization. Examples of algorithms are the ones proposed by McCormick (1976). Floudas and Viswewaran (1990) and Sherali and Alameddine (1992) which can be used to solve bilinear programming problems like the ones that arise in networks with multicomponent streams. For a recent review in the area of bilinear programming see Al-Khayyal (1992).

As for previous work in the design and synthesis of multicomponent process networks Mahalec and Motard (1977) and Nath (1977) developed evolutionary techniques that are based on heuristics to generate a network configuration. Floudas (1987) addressed the synthesis of separation networks with mixed products in which only sharp separators are considered. A superstructure of the process network was proposed and modelled in terms of concentrations. The resulting model is nonconvex and solved with a standard NLP algorithm with no guarantee of global optimality. Floudas and Aggarwal (1990) solved small pooling and blending problems and sharp separation networks problems using a strategy based on Benders decomposition. In this approach only convex subproblems are solved but there is no guarantee of obtaining the global optimum. Kocis and Grossmann (1989) modelled process networks with multicomponent streams in terms of the individual component flows. They included a set of bounding constraints with binaxy variables to approximate the nonconvexities that are present in splitters with multiple outputs. Wehe and Westerberg (1987) studied the problem of sharp separation networks with mixed products. They proposed a search procedure that involves the

enumeration of the different separation sequences. The nonconvex equations are dropped and constraints that are valid for each particular sequence with a set of bounds over the key components are included to obtain tighter UP relaxations for each configuration. However, the number of sequences to be examined grows rapidly and there is no guarantee of global optimality.

In some particular cases the nonconvexities in the mass balances can be avoided through the introduction of binary variables. One of these cases is when single choice splitters are present in the flowsheet (Kocis and Grossmann, 1989). Here, it is possible to have a mixed integer linear formulation for the mass balance equations in terms of the individual component flows. Another restricted case for which some nonconvexities can be reformulated is when mixing within the network is only allowed for streams of the same concentration. In this form, larger network superestructures must be proposed and the concentrations of the streams are known beforehand. Integer variables are introduced to model the existence of the different streams (e.g. the mixed integer linear formulation for sharp separation networks by Floudas and Anastasiadis, 1988).

The objective of this paper is to present an efficient global optimization method that exploits the particular structure that is present in process networks with multicomponent streams (e.g. pooling and blending systems, sharp separation networks). First a relation is established between formulations based on concentrations and individual flows. This is done following the Reformulation-Linearization technique proposed by Sherali and Alameddine (1992). As will be shown, a linear relaxation Is obtained that is in the space of the concentrations and individual flows which can be used in a branch and bound search is to find the global optimum. Application to the optimal design of sharp separation systems with single feed and mixed products is considered In detail. Different preprocessing techniques that allow tightening of the relaxation problem are presented. The performance of the algorithm is reported on a total of twelve problems.

Modeling with concentrations and individual flows

Consider a process network that consists of splitters, mixers and process units that are interconnected with multicomponent streams (see Fig. 1). The process iinits that are considered in this paper are units in which the output flows of the components can be expressed as a linear relation of the inlet flows (e.g. sharp separators, reactor with^known conversion). It is possible to formulate the mathematical model of the process network in terms of the concentrations of the streams. Xj^k. Another possibility is to model the network

using flows of individual components. The former has the advantages that it provides a convenient framework for the evaluation of thermodynamic properties, and in many cases bounds can be expressed in a more natural form. A major disadvantage is that many nonconvex terms (bilinear) are involved in the mass balances for the components. The individual component flows formulation is often chosen since it gives rise to a larger number of linear equations and the only nonconvexities are involved in the modelling of the splitters. In these untis it is necessary to enforce that the components maintain the same concentration in each of the streams leaving the splitter. These constraints can be expressed as relations between the different components (Wehe and Westerberg, 1987). One deficiency of this representation is that since many flows can take values of zero, singularities may arise with which conventional nonlinear programming methods may have difficulties to converge. Another alternative is to introduce additional variables that represent split fractions (Kocis and Grossmann. 1989). This involves a larger number of constraints but tends to yield a formulation that numerically is better behaved.

Following are the equations that apply to the mixers, splitters and units using the two alternative representations:

Mixer

A mixer k consists of a set of Inlet streams, M_k, and an outlet stream k (see Fig.2).

a) Concentrations

The total mass balance for a mixer k is given by:

$$F^k = \sum_{i \in M_k} F^i \qquad (1)$$

where F^i is the total flow in stream i. The mass balance for each component j is given by the nonlinear equations.

$$F^k x_j^k = \sum_{i \in M_k} F^i x_j^i \qquad \text{for all } j \qquad (2)$$

where x_j^i is the concentration of component j in the stream i

b) Individual flows

Here it is only necessary to write a mass balance for each component j, given by the linear equations:

$$f_j^k = \sum_{i \in M_k} f_j^i \qquad \text{for all } j \qquad (3)$$

where f_j^i is the flow of component j In stream i.

Splitter

A splitter k has an inlet stream k and a set of outlet streams S_k (see Fig. 3).

ql[Concentrations

The equations for a splitter in terms of the concentrations are given by the following linear equations

I ' P = P̂ * **(4)**
1 € Sfc

Xj* = x,^{k} **for** **all i 6 S_k a$dj** **(5)**

1 x ^ = 1 **" (6)**
J

b) Individual_flows

The mass balance for each component j is given by

If ^ a f A **for all j** **(7)**
t€Sfc

Here, it is also necessary to enforce the condition that the streams leaving the splitter have the same proportions in flow for each component. These relations between components, which are nonlinear, can be expressed in terms of the inlet stream k and a given component j'

f_r^k fj* s fj^k fjJ **for all i € I_k and j * j^1** **(8)**

A different approach consists of introducing as additional variables the split ratios $§^f_t$ that represent the part of the inlet flow that goes to the outlet stream i. The nonlinear equations are given by

ff«9fj^k **for alii € I_k and j** **(9)**

withO£?£l.

<u>**Process units**</u>

In this paper it will be assumed that the outlet streams, i € O_k, in the process units can be expressed as linear relations of the inlet streams, i e I_k (see Fig. 4). This is for instance the case of sharp split separators, separations in which the recovery level is known, or reactors that have a fixed conversion.

a) Concentrations

The overall mass balance for process unit k is given by,

I Pa I P **(10)**
1€I_k i€O_k

The mass balance for each component j is given by the nonlinear equations;

∑ βjr'lk F' xj' = Px,^{4} **for all ie O_k and j** **(11)**
i' € I_k

where P_{jr}^{lk} is a constant for process unit k that gives the distribution of component j in the stream i e O_k coming from streams i^1 *e* I_k. For a separator unit it is required tliat 2 IV* = 1 (i € O_k) and £ P_{jr}^{tk} a 1 (i' € I_k). A sharp split separator is one for which II_kl = 1 and IO_kl =.2 (top and bottom streams) and for all the components the constant p_{jr}^{ik} are either 0 or 1.

b) Individual flows

Only the mas balance for each component is necessary and it is given by:

$$f_j^i = \sum_{i' \in I_k} \beta_{ji'}^{ik} f_j^{i'} \qquad \text{for all } i \in O_k \text{ and } j \qquad (12)$$

A model in terms of individual flows MF consists of the linear equations (3), (7) and (12) plus the nonlinear equations (8) or (9). The model in terms of the concentrations, MX, includes the linear equations (1), (4), (5), (6) and (10) plus the nonlinear equations (2) and (11).

Reformulation and Linearization

In order to avoid the direct use of the nonconvex models MX and MF, there is a relation that can be established between them using the reformulation and linearization technique for bilinear programming models proposed by Sherali and Alameddine (1992). This technique can be applied to the model MX. First, consider the bounds over the variables present in the bilinear terms (total flow, F^i and concentrations x_j^i)

$$F^{iL} \le F^i \le F^{iU} \qquad (13)$$

$$x_j^{iL} \le x_j^i \le x_j^{iU} \qquad (14)$$

Using the bounds in (13), (14) the following constraints can be generated for the bilinear terms in (2) and (11),

$$F^i x_j^i \ge F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

$$F^i x_j^i \ge F^{iU} x_j^i + x_j^{iU} F^i - F^{iU} x_j^{iU} \qquad (16)$$

$$F^i x_j^i \le F^{iU} x_j^i + x_j^{iL} F^i - F^{iU} x_j^{iL} \qquad (17)$$

$$F^i x_j^i \le F^{iL} x_j^i + x_j^{iU} F^i - F^{iL} x_j^{iU} \qquad (18)$$

In fact, McConnick (1976) has shown that the constraints in (15)-(18) correspond to the convex and concave envelopes of the bilinear terms over the given bounds. The formulation is linearized by the definition of the following variables:

$$f_j^i = F^i x_j^i \qquad (19)$$

Tile resulting model which involves equations UK (3). (4). (5), (6). (10). (12) and the constraints in (15M18) is a linear relaxation of the original nonconvex concentration model, MX, in which the nonlinear equations (2) and (11) have been replaced by the linear equations (3) and (12) from the individual flow model. MF. It is possible to generate additional linear constraints that are redundant to the original nonlinear model, MX, but that can be nonredundant in the linear relaxation of the model (Sherali and Alameddine, 1992: Sherali et

al*. 1992). In particular, consider equation (7) that is the linear component mass balance for the splitters in model MF. "This linear equation is not present in the linear relaxation of the concentration model. MX. Take equation (4) and multiply by the valid bound constraint x_i^k £ 0 to get

$$\text{£}_{i \in S_k} F^* x_J^k = F^k \wedge^k \qquad (20)$$

Using equation (5) yields.

$$\text{£}'_{i \in S_k} F^{**} i^l \text{»} lE^* \wedge^k \qquad (21)$$

that can be linearized to.

$$\sum_{i \in S_k} f_j^i = f_j^k. \qquad (22)$$

yielding equation (7). Hence, the linear equation for the splitter is valid and it is included. The nonlinear equations (8) or (9) can also be generated in a similar fashion but their linearizations are in general redundant (see Appendix A). They are only useful when the formulation 6f the problem provides non-trivial bounds over certain components in the outlet streams of a splitter, or when there exist some restrictions over the split ratios for the outlet streams.

Also, the constraints that relate the total flow and the individual flows of a stream can be generated for the splitters. Taking equation (6) and multiply by F^1 yields.

$$F^i \sum_j x_j^k = F^k \qquad (23)$$

Using the constraints $x_i^1 = x^{\wedge k}$ in equation (23) and linearizing with fj^1« $F^* x_i^4$ yields,

$$F^i = \sum_j f_j^i \qquad (24)$$

Based on the above it is possible to obtain a reformulated model MR that involves concentrations, total flows and component flows, and which bounds the solution of the original problem. The following equations are given for model MR:

a) Objective function. <t: which is expressed in terms of individual or total flows.

b) Mixer equations, which are expressed in terms of the total and individual component flows.

$$\bullet.F^* \text{«} \ \text{£}_{t \in M_k} F^* \qquad (1)$$

$$f_j^k = \text{If/}_{i \in M_k} \qquad \text{forallj} \qquad (3)$$

c) Splitter equations, that are expressed in terms of the individual component flows and the f^oncditi^ tf^nff of the streams

y p-pk (4)
t€S_k

Xj' = Xj^k for alii e 3* and J (5)

XxJ<=i (6)

£ If «ff for ail j (7)

d) Process units equations, that are given in terms of the total and individual component flows

$\sum_{i \in I_k} F^i = \sum_{i \in O_k} F^i$ (10)

fj«= T p/'f/ foraUie $O_k$andj (12)

e) Relation between the total flow and the individual component flows

$F^i = \sum_j f_j^i$ for all streams (24)

f) Linear estimators, relate the individual component flows with the total flow and concentrations.

ri £> piLjqi + x* F* pi . piL jri L (15)

fji ^F^1tt^4^^!^uF^l-F^lu^lu for alii e S_k (16)

f/ ≤F^ltt^l4-^1LF^i-F^!u^IL i s k € splitters flT)

fj^1 ^F^XJ' + X^F-F'-XJ'" foralljeJ (18)

f) Bounds on flows and concentrations

$F^{iL} \leq F^i \leq F^{iu}$ (13)

$x_j^{iL} \leq x_j^i \leq x_j^{iu}$ (14)

In previous approaches (Wehe and Westerberg, 1987; Kocis and Grossmann, 1989) looser approximations of the nonlinear terms were used. In both cases, the nonconvex problem (MF) was relaxed to a linear model by dropping the nonlinear equations (8) or (9). Equations that approximate the difference relation between the components were considered (Kocis and Grossmann, 1989). They were based on the difference that exists at the inlet of the splitter between the flowrate of the components and required the introduction of binaiy variables.

Outline of global optimization method

Model MR can be applied to predict lower bounds to the global optimum in the optimization of pooling and blending problems and in the synthesis of separation systems. The reason is that model MR provides a valid relaxation of the original feasible region since the nonlinear equations (2) and (11) in model MX are not considered, and the valid linear equations (3), (7), (12) and (15)-(18) are included. The proposed global optimization algorithm relies on the solution of the relaxed problem MR within a spatial branch and bound enumeration. The outline of the algorithm is as follows (for a more detailed description of step 4 see Quesada and Grossmann (1993))

0. Preprocessing (optional)

Determine bounds on the variables involved in the nonconvex terms, that is total flows, F^1, and concentrations, $x\{$. Apply any additional preprocessing specific to the structure of the problem in order to further bound or flx variables.

1. *Lower Bound*

Solve model MR over a given subregion (initial subregion is the complete feasible region) minimizing a convex objective function $. If ❖ is linear the model Is an LP.

2. Upper Bound

Any feasible solution to the nonlinear model provides an upper bound. Heuristic techniques can be employed to obtain good feasible solutions or the original problem, MF. can be solved using the solution of model MR as a good initial point. If the solution of problem MR is feasible it provides an upper bound.

3. *Convergence*

If the lower bound of a subregion is sufficiently close or above the upper bound discard that subregion. If no subregions are left the global solution corresponds to the best upper bound.

4. Branch and Bound

Partition the remaining subregions into a set of disjoint subproblems. Repeat steps 1-3 for each of the new subregions.

Remarks

The preprocessing step plays an important role in the above algorithm. It is during this step that initial bounds for the variables involved in nonconvex terms are obtained. The quality of

these bounds affects the tigthness of the lower bound since they are part of the estimator equations (15)418). Additionally, these bounds affect the performance of the algorithm because they define the search space over which the branch and bound procedure may have to be conducted.

In some cases, as described later in this papier, it is possible to exploit the particular structure of the process network and generate bounds for the variables without having to solve any subproblems. Furthermore, during this preprocessing step additional constraints can be generated for predicting a tighter lower bound of the global optimum can be obtained.

Some of the linear mass balances and the estimator equations are redundant in the nolinear formulations, MF and MX. These equations become nonredundant in the reformulated model, MR. and for that reason it is important to write a complete formulation of the network. However, this model can present some redundancies that can be easily identified and eliminated to reduce the size of the model. This is the case for the concentration variables used in the splitters. Model MR uses different sets of concentrations variables for the inlet and outlet variables of a split unit. In practice, it is only necessary to define the concentration of the component in the splitter and use the same variables for all the splitter streams. Also, some redundancies can occur with the total flow variables. These ones are necessary for the streams in the splitters but they might be redundant and eliminated in the other untis if they do not appear in other part of the model or in the objective function.

If the solution of model MR is feasible for the original nonconvex problem then it corresponds to the global optimal solution. When the solution to the model MR Is not feasible it is necessary to follow a blanch and bound procedure to search for the global optimum. This procedure requires a valid upper bound on the global optimum. This can be generated through heuristics or by solving directly the nonconvex model. For this purpose, the process network model is formulated in terms of the individual component flows and the nonconvex equations for the splitters are included. Equation (9) was also used in this work to model the splitters due to it is better numerical behavior. The solution to the model MR was used for the good initial point. In many instances, it was not possible to solve these nonlinear problems with MINOS 5.2. The nonlinear models were solved using CONOPT in GAMS 2.25.

During the branch and bound procedure a tree search is generated. Of the set of open nodes, these are the nodes that have a lower bound that is e-smaller than the current upper bound, the node with the smaller upper bound is selected to branch on. The splitter units are the units that are approximated, and of these, the splitter that has the largest difference

between its approximated and actual individual component flows is selected. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. The branching is done in the selected splitter over the concentration of the component that has the largest difference.

First, the actual concentrations for the individual component flaws in the LP solution (*) for the splitters are calculated by,

$z_j^k = \frac{l_j}{F^{k}}$ for all the inlet streams to splitter (26)

The splitter unit m is then selected according to the equation.

$m = \arg\max_k$ c mitt ($\sum_{S_k} \sum_{j=1}^{N}$ | ty-z^FH) , (27)

Equation (27) represents the total difference between the LP solution for the flows after the splitter and the actual value of these flows considering the concentrations before the splitter. Once the splitter has been selected, the component in that splitter that has the largest difference. J\ is selected by,

J--aigma^(JL"I f/ - z»^{k} F* 11 $_{S_m}$ (28)

The following branching constraints are then used;

$x_j^m \le z_j^m$ and $x_j^m \ge z_j^m$ (29)

To improve the upper bound it might be necessary to solve additional nonconvex problems. These can be solved after a given number of nodes using the solution of the node with the smallest upper bound as the initial point. In this work if there was no significant change in the lower bound of the new nodes with respect to the lower bound of the parent node (< 1%) a new nonconvex problem was solved.

Example 1

Consider the following pooling and blending problem by Harveley (1978). Two streams that have components A and B are mixed in a initial mixer a then go through a splitter to obtain two streams than can be mixed with an additional stream (see Fig. 5a). Two different products can be obatined and there are constraints on the concentration of component A in these products. The objective function consists of niimizing the cost that is given by the total flow of the streams times the cost coefficients, c_t, given in Fig. 5.

This problem has two local solutions. One has an objective function 0 = -100 and consists of only producing product 2. The other local solution, that corresponds to the global optimum, has an objective of + « -400 and here only product 1 is produced.

Model MR is formulated for this problem and the initial lower bound is 0_L = -500. The nonlinear model. MF, is solved using the solution of model MR as the initial point and an upper bound of $ s -400 is obtained. Since there is a gap between the bounds of the global solution a partition is performed. There is only one splitter that needs to be approximated and since there axe only two components it is irrelevant which one is selected since the composition bounds are related (eg. $x_B^L = 1 \bullet x^{*u}$). The actual value of the composition of A in the solution of model MR is used as the branching point (x* = 0.0166) to generate two new subproblexns. The first subproblcm (x_A £ 0.0166) has a lower bound of 4_L = -100 and the second subproblem (x* £ 0.0166) has a solution of fa. = -400 (see Fig. Sb). Both of these bounds are greater or equal than the upper bound, therefore the global solution has been found (x_A = 0.01).

Example 2

The next example is a separation problem taken from Kocis and Grossmann (1987). The original problem has binary variables in the formulation and they have been fixed to 1 for this example (see Fig. 6).

The top stream of the flash unit has 85% of the inlet flow of A and the bottom stream has 80% of the inlet flow of component B. In the column. 97.5% of the inlet flow of A goes to the top whereas 95% of the inlet flow of B goes to the bottom stream. Hie total flow to the flash unit and the column have to be greater than 2.5 and smaller than 25, whereas the total flow of each of the two feed streams has to be less than 25. The objective function is given by,

$$\Phi = 52 + 10 F! + 8 F_2 + F_4 + 4 F_5 - 35 Pj^A - 30 P_2^B \quad (30)$$

The initial lower bound for this problem is 0_L = -513.22 and it is infeasible for the original NLP model. A nonconvex problem is solved using CONOPT with the solution of model MR as the initial point obtaining an upper bound of Φ = -511.87 and the relative gap is only 0.3%. Again only one splitter is present in the network and a partition can be performed using the concentration of component A in this splitter. The lower bounds for the new two subproblexns are *fa* = -511.87 (x_A £ 0.5121) and Φ_L = -511.80 ($x_A \geq$ 0.5121). Both solutions are greater or equal than the upper bound and the global solution has been obtained. In the global solution Fi = 8 and F_2 = 25, and 11% of the inlet flow lo the splitter is directed to the flash. 76% to the column and the rest bypassed to P_2.

Example 3

TIUs example corresponds to a separation problem with three feeds and three product streams. The network configuration and product specifications are given in Fig. 7. The objective function is given by

$$>\ll 4Si + 1.5S2 + 4S3 \qquad (3D$$

The initial lower bound is 4_L = 138.18 and the nonconvex problem MP is solved obtaining a solution of $ = 138.7. The gap between these bounds is less than 0.4 %. The global optimum for this tolerance is shown in Fig. 8.

Sharp separation networks

In order to illustrate the application of the above algorithm to a specialized case where the structure can be further exploited, the problem that will be considered is the synthesis of separation networks with single feed and mixed products that consist of sharp separators and bypasses. It is assumed that a single feed with N components must be separated into K specified multicomponent product streams. The components are ordered from the lightest to the heaviest.

A modification of the superstructure proposed by Floudas (1987) for this problem is used (see Fig. (9)). The superestructure consist of N-l separators. Separator i performs the task of removing component number 1 to number! at the top of the separator and components number i+1 to N at the bottom of the separator. The feed to the network is split into N-l streams, F_t, that go to the separators and K streams, a_k. that bypass the network to go to the products. Each stream $F_{\{}$ is mixed before the separator i with streams that come from the top and bottom streams from the other separators to obtain the inlet stream to separator i, S^

The outlet streams of separator i are the top, $T_{(}$, and the bottom, B_t. These streams, T, and B_t. are each split into streams, PT_t^k and PB^ respectively, that go to the K products and into streams, KIV' and.RBf. that are redirected to the other separators. The top stream of separator i, T_{it} can be redirected only to the separators 1 to i-1 since it can only contain components number 1 to i. It would not be optimal to send part of this stream to any separator from i+1 to N since no separation would be achieved and a bypass of these separators would achieve the same separation with smaller flows. *KTf* is the flow redirected from the top of separator i to separator i\ In the same fashion that with the top stream, the bottom of separator i, B,, can be redirected only to separators i+1 to N-l since it can only

contain components that are separated by these sharp separators. RB/ is the redirected flow from the bottom stream of separator i to separator i\

Model

It will be assumed that the objective function can be expressed as a linear function that depends on the total flow to each separator. The model expressed in terms of concentrations and total flows has the following form:

min •= Z c t S | (N-1, t=1) (32.1)

st. reeo!•» 2u *i * m®k (N-l, t»i; K, k*i) IOZ.ZJ

L sF|L **for all i and j** **(32.3)**

Si» F| + £ RB,[1] + X Kiy (1-1, t'«l; N-l, !'«*•!) for aU i (32.4)

Si xsq = flj +t*Q RBf[1] x^j + t*£•KTr! xtrj (1-1; N-l) **for all i and j** **(32.5)**

S i s ^ + Bi **for alii** **(32.61**

Ti xtij = Si xsij **for all i and ord(j) ≤ i** **(32.7)**

xtij = O **for aU i and ord(j) > i** **(32.8)**

Bi xbij = Si xsij **for all i and ord(j) > i** **(32.9)**

xbij = 0 **for all i and ordQ) ^ i** **(32.10)**

Tf m £RTi[r] + £FTt[k] (t-i, 1*1; K, k«l) for aU i (32.11)

Bi s t*£RB^f + k£PBt[k] (N-l; K) **for all i** **(32.12)**

Pk = ∑PTi[k] + ∑PI3^ + 0^ (N-l, i=1; N-l, i=1) Tor'alt k (32.13)

Pkj = £PT,[k] xt«j + £PB,[k] *xbq*+ a_k z, (N-l, i*l; N-l, i»l) for all k and j (32.14)

∑ xtij = 1 (i, ord(j)=1) for alii (32.15)

Z xb,j = 1 (N, ord(j)=i+1) **for all i** **(32.16)**

I x s , j = l (N, ord(j)=i) **for alii** **(32.17)**

Si, T|t B,. Fi. RT/. RBi*'. PTt[k]. PBi[k]. c*. *t*^ xs,j, xty. xbij ≥ 0

The parameters Feed, zj, P^k and p_{kj} represent the total feed, composition of the feed, total flow of product k and component flow of component j in product k, respectively. The variables xsq, xty and xbg are the concentrations of component j in the inlet stream to separator 1, top of separator 1 and bottom of separator i, respectively.

The objective function (32.1) is a linear function of the inlet flow to the separators. Equation (32.2) is the total mass balance in the inlet splitter and equation (32.3) is the component mass balance. Equations (32.4) and (32.5) are the total and component mass balances for the mixer i before the separator L The material balances for separator i are given by equations (32.6)* that is the total mass balance for the separator, equations (32.7) and (32.8) that are the component balances for the top stream and indicate that nothing from components number 1+1 to N-l is in the top of the separator, and equations (32.9) and (32.10) that are the component mass balances for the bottom streams. Equations (32.11) and (32.12) are the overall mass balances for the splitters of the top and bottom streams alter separator 1. The equations that state that the concentrations of the outlet streams should be the same that the inlet stream in a splitter have been already substituted. Finally, equations (32.13) and (32.14) are the overall and component mass balances for the mixer for product k.

Model (32) corresponds to a formulation of the type of model MX where the distribution coefficients are known and restricted to 0 or 1. Some simplifications have been made to avoid including many irrelevant variables (e.g. not to define concentrations for the streams that go the top i to product k). Although, some of the linear constraints in this formulation are redundant, they can become nonredundant in the linear relaxation as will be shown in Example 4.

Equations (32.5). (32.7). (32.9) and (32.14) involve nonconvex terms. This model can be reformulated as in model MR by introducing individual component flows and the linear equations (15M18) and (7) according to the approach illustrated earlier to obtain a model in the form of model MR The resulting reformulated model is as follows,

$$\min \phi = \sum_{i=1}^{N-1} c_i S_i \qquad (33.1)$$

$$\text{st} \quad Feed = \sum_{i=1}^{N-1} F_i + \sum_{k=1}^{K} X_k \qquad (33.2)$$

$$f_{ij} = F_i z_j \qquad \text{for all i and j} \qquad (33.3)$$

$$S_i = F_i + \sum_{t=1}^{i-1} RB_t^i + \sum_{t=1}^{N-1} RT_t^i \qquad \text{for all i} \qquad (33.4)$$

$$s_{ij} = f_{ij} + \sum^{i-1} rbt_j^i + \sum_{i=t+1}^{N-1} rt_{rj} \qquad \text{for all i and j} \qquad (33.5)$$

Sr-TV+'B, for alii (33.6)

t_n = sij for all i and ord(jJ £ i (33.7)

t_f * 0 for all i and ord(j) > i (33.8)

$b_{,jSS}$Sg for all i and ord(j) > i (33.9)

t,j = O for all i and ordfl) £ i (33.10)

$T, = \sum_{1*1}^{t} sW + \sum_{k«l}^{K} lPT_4^k$ for all i (33.11)

$B_i\ m\ \sum_{i'=i+1}^{N-1} £RB,^{if} + \sum_{k=1}^{K} £PB_t^k$ for all i (33.12)

$p^* = \sum_{i=1}^{N-1} TFT_t^{k''} + \sum_{i=1}^{N-1} £PB|^k$ + otk for all k (33.13)

$Pki * \sum_{i=1}^{>M} 5jtt,^k + \sum_{i=1}^{riti} Xpb\wedge^{k+}$ ctk z, for all k and j (33.14)

$t« = \sum_{i'»l}^{1*1} Z \wedge / + \sum_{k«l}^{K} Zptij^k$ foraUiandj (33.15)

$b« = \sum_{l'*t+l}^{N-l} Zrbij^{4'} + \sum_{k»l}^{K} £pb«^k$ for all i and j (33.16)

$\sum_{ord(j)=I}^{i} xt_{ij} = 1$ for alii (33.17)

$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1$ for all i (33.18)

$T_i = \sum_{ord(j)=1}^{i} t_{ij}$ for alii (33.19)

$B,s\ \sum_{ord(j)=i+1}^{N} Ib«$ for alii (33.20)

$PTi^k = \sum_{oixi(j)»i'}^{i} Tpt<*^k$ for alii and k (33.21)

$PB,^k = \sum_{ord(j)=i+1}^{N} Ipbq^k$ for alii and k (33.22)

$RT/ = \sum_{ord(i)-l}^{i}]DV$ for all i and $i^f < i$ (33.23)

$RB_i^{f} = \sum_{ord(j)=i+1}^{N} Irb/$ for all i and r > i (33.24)

Equations (15-18) for t_{4J}, rt_{tJ}^{l} and pt_{tj}^{k} in terms of xt_{ij} and the total flow of its respective stream.

Equations (15-18) for bq, rbq* and pb_{fj}^{k} in tenns of xb_{ij} and the total flow of its respective stream.

S_i. T_{lt} Bt. F_4. KTf, $RB,^{1'}$, $PT,^k$. PB^. a_k. f,j. $xs,_{jf}$ $xt4_{jf}$ xb,j ^ 0

s_{ij}, t_{ij}, b_{ij} pt_{ij}^{k}, pb_j^{k}. rt,/. $rby^{1}*$ ≥ 0

It is not necessary to include equations (15)-(18) for the inlet component flows to the separator, Sq. since the variables xsy only appear in these equations. Also, the component flows, S|j9 only appear in mixers and sharp separators units which can be exactly represented in terms of the individual component flow equations (33.5). (33.7) and (33.9). Equations (33.15)-(33,16) that are the component mass balances for the splitters of top and bottom streams have been included accordingly to the reformulation previously presented. Equations (33.19M33.24) relate the total flow and the individual component flows for the splitter streams.

Preprocessing

The proposed Superstructure (Fig. 9) allows to bypass certain amount of the feed to the product k. Ok, without having to go through the separation network. The amount of the product k that is not bypassed has to be processed In the separation network and it will be denoted as the 'residual product*. Hence, the total 'residual product k^f is given by (P^k - a_k) and has the component flows given by (p^ - o^Zj)(see Fig. 10).

The global optimal solution of model (32) is a network in which all the 'residual products[1], have at least one component with a zero flow. The reason that it is not optimal to separate a stream in the network and later to remix it. The same degree of separation can be achieved using a bypass that does not incur any cost in the objective function.

Consider the second separator in the solution obtained by Floudas (1987) to his second example (see Fig. 11). For this subnetwork of the complete structure the "upper 'residual product' has components B and C present. The components are being separated and remixed again. The same outlet flows can be obtained with a smaller input flow to the separator as it is shown in Fig. 11. Note that both 'residual products' have components with zero flow.

It should be clear that if there was not a component with zero flow in the 'residual product[1], then there is part of this stream that could have been obtained by just bypassing the network. This in turn does not incur in any cost, whereas going through the network has a positive cost. The above condition gives a lower bound for the bypass to each product. This also corresponds to the largest amount that can be bypassed since all this flows in the 'residual flow[1] have to be positive. In this form the bypass can be precalculated without affecting the global optimality of the solution.

The bypass to product k is given by the maximum amount that can be sent to product k without having a negative flow; that is.

$$a^* = \min_j t \,^\wedge 4 \qquad (34)$$

where Zjis the feed composition and p_{kJ} is the flow of component j in product k. The component flews for 'residual product[1] k, $y\#$ are given by.

$$\gamma_{kj} = p_{kj} - \alpha_k z_j \qquad (35)$$

Key component bounds

Wehe and Westerberg (1987) proposed using lower bounds for the flow of the key components in separator i. These bounds are based on the fact that separator i is the only unit that can perform the task of separating component number i from component nutaber i+1. They are redundant for the nonlinear formulation (32) but they are relevant for the linear relaxation in (33). To calculate them, it is necessary to determine in each product what is the difference between the two key components of separator 1 with respect to the concentrations in the feed. The lower bounds in separator i* for the flow of the key components in the top (component Jl) and bottom (component j2) streams are given by:

$$b_{tj1} \geq \sum_{k=1}^{K} \{ \gamma_{kj1} - z_{j1} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (36)$$

$$b_{bj2} \geq \sum_{k=1}^{K} \{ \gamma_{kj2} - z_{j2} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{foralli} \qquad (37)$$

where y^ is given by (35). It is important to include both bounds in the relax model (33) since there is no guarantee that the inlet flow to separator i has the same proportion between the key components as the feed. It is not known in which part of the sequence separator 1 will be placed, and it can be after a splitter that is not being approximated correctly.

The bounds in (36) and (37) can be extended to separation of components that are not adjacent in the feed. Consider component number i and component number i+3. There are three separators that can perform this task, separators i. i+1 and i+2. Cuts of the following form can be obtained,

$$b_{t\,i} + b_{t\,i+1} + b_{t\,i+2} \geq \sum_{k=1}^{K} \{ \gamma_{ki} - z_i \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (38)$$

$$b_{b\,i} + b_{b\,i+1} + b_{b\,i+2} \geq \sum_{k=1}^{K} \{ \gamma_{k\,i+3} - z_{i+3} \min_{ord(j)=i \text{ or } i+3} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for all i} \qquad (39)$$

Equations as the ones in (38) and (39) can be redundant compared to equations (36) and (37) and it is possible to detect this before solving the problem.

Relative flowrate constraints

These constraints are used when the relation between the flowrates of two components is known. In particular, consider component A in the last column of the network (we Fig. 12). None of the redirected streams contains component A. Therefore, the relative flowrate of component A with respect to the other components in the top stream has to be smaller than in the feed. This relation should remain valid after the top stream is split to the products and redirected flows.

In the separator previous to the last one, N-2, all the streams do not have component A except the one coming from the top of the last column. For this one it is already known that the relative flow of component A with respect to the other components is smaller than in the feed. This type of analysis can be done for component A and component N in all the columns yielding th$ following linear constraints for the splitters.

for alii and k

ZAPtij^{k}-ZjPt«A^{k}^O for j * A and ord(j) ≤ i (40)

z_N pb,j^{k} - z, pt*$_N^{k}$ £ 0 for ord(j) * N and ord(j) > i (41)

for all i and i' > i

ZArt|j^{r}•ZjrtiA$^{4'}$^0 for J * A and ord W ^ * W2)

for alii and f < i

z_N rb$_{ij}^{r}$ - z_j rb$_{iN}^{r}$ ≥ O for ord(J)*W and ord(j)>i I43J

Boundson concentrationsand total flows

The approximations (15-18) require bounds for the total flows and component concentrations in the splitters. The lower bound for the total flow of the top and bottom streams is given by the lower bound of the key components obtained in equations (36) and (37). For the outlet streams of the splittters, that are the redirected streams and the streams that go to the products, the lower bound is zero. The upper bound for the total flow of the top and bottom streams is given by the feed to the network of the components that can be present in each stream .that is,

$$T_t^u = \sum_{ord(j)=1}^{t} (Feed - Xak) z, \qquad \text{for all } i \qquad (44)$$

$$B|« = \sum_{ord(j)=i+1}^{N} t Feed - IOfcJ z, \qquad \text{for all } j \qquad (45)$$

The upper bound for the streams after the splitter are given by.

$$RT_i^{i'u} = T_i^u \quad \text{for ail i and i' < i} \quad (46)$$

$$RB_i^{i'u} = B_i^u \quad \text{for all i and i'> i} \quad (47)$$

$$PT_i^k = \sum_{ord(j)=1}^{i} \gamma_{kj} \quad \text{for all i and } k \quad (48)$$

$$PB_i^k = \sum_{ord(j)=i+1}^{N} \gamma_{kj} \quad \text{for alii and k} \quad (49)$$

The lower bounds for the concentrations are zero except for the key components in the separator for which the lower bounds are given by the lower bound of its flow divided by the upper bound of the total flow of that stream. The upper bounds in the concentrations are given by one minus the lower bounds of the other components.

The solution of the linear programming model (33) provides a lower bound to the global optimum since this model is a valid relaxation of the nonconvex model (32). This lower bound is obtained by solving the LP model for the 'residual products' in 135) with the addition of the valid constraints (36)-(43).

The upper bounds are generated solving mode} (32) in terms of the individual flows for the 'residual products'. When additional nonconvex problems are solved to improve the current upper bound it can happen that very similar initial point are generated. In this case, a new nonconvex NLP is solved in which bounds over the total inlet flows to the separators (SJ are included. For this purpose the values of these variables in the LP solution (S_4*) are used such that the current incumbent solution is no longer feasible.

Example4

Consider the 3 component example proposed by Floudas and Agganval (1990). An equimolar feed has to be separated into two products as show in Fig. 13. The objective function is given by

$$\Phi = 0.2395 + 0.00432\ S, + 0.7584 + 0.01517\ \&* \quad (50)$$

The bypass to products 1 and 2 can be calculated according to equation (34) and the 'residual product' component flows are obtained through equation (35) (see Fig. 14). The total bypass to product 1 is a, = 90 and the bypass to product 2 is ct_2 = ISO and the feed has a concentration of z_A = 1/3, z_Q = 1/3 and Zc = 1/3. In this form the 'residual product' 1 is YIA = 0, YiB = 20 and y_{lc} = 0 and the residual product' 2 is Y_2A = 20, Y_{2B} = 0 and Y2C = 20. Additionally, lower bounds on the flow of the key components in both separators are obtained

using equations (35)-(36). The key components in separator 1 are component A in the top and its flow has to be at least 20 and component B in the bottom has to have at least a flow of 20. In the top stream of the second separator at least 20 units of component B have to be separated from 20 units of component C in the bottom stream. It is important to note that after preprocessing the network several suboptimal solutions have been cut off. One of these suboptimal solutions for this particular data is a parallel configuration of both separators (there are situations in which a parallel configuration can correspond to the global solution as will be shown in example 5). In this example the direct or indirect sequence have a lower objective function. Both of these configurations are local solutions with an objective function value of 4 » 1.8639 for the direct sequence and 4 = 2.081 for the indirect one. In some instances, MINOS 5.2 had problems converging even in this small example.

The LP (33) is formulated for this problem, giving a lower bound of (^ = 1.8639. The approximations are exact and therefore this solution is a feasible solution of model (32) proving that it corresponds to the global optimum. Hence, convergence is achieved in one iteration. The optimum solution corresponds to the direct sequence shown in Fig. 15. It should be noted that if the linear mass balances for the mixer for product 2 were not considered since they are redundant for the nonlinear formulation, a lower bound in the relaxed model of Φ_L = 1.12 is obtained, this shows that it is relevant to include all the mass balances in the linear model in order to tighten the lower bound.

Example 5

In the approach proposed by Wehe and Westerberg (1987) for the case of 3 components only the direct and indirect sequences are considered and both options can be modelled as LP problems since no mixing is required for these separation networks. However, this example shows that parallel configurations can be also globally optimal and that they are not excluded by the method proposed in this paper. To be able to consider parallel configurations or any combination of parallel with direct or indirect sequences it is necessary to model a superstructure in which mixing is allowed (like in the structure used in Fig. 13). Here, nonconvexities arise in the mass balance equations after the separators.

Consider that an equimolar feed is to be separated into the two different products given in Fig. 16. The objective function is to minimize the sum of the total flows into the separators. The same procedure that in the previous example is followed and the bypass can be precalculated by equation (34). The solution to the model (32) yields Φ_L = 12 and since it is a feasible solution of model (32) it corresponds to the global optimum (see Fig. 16). Note that the solutions for the direct or indirect sequences have an objective function of Φ =16.

Branch and Bound

If there is a gap between the lower and upper bound a branch and bound search is performed. It is only necessary to do the search over the variables involved in the nonconvex terms. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter* In this way. it is necessary to check the approximation for the concentrations in the splitters of the top and bottom streains of the separator. Equations (26)-(29) for the splitters of top and bottom streams are used to perfom the branch and bound search.

Results

Table 1 summarizes the results of the earlier examples 1 to 3 and of the sharp separation network examples 4 to 12. The number of variables is the total number of variables that are included in the reformulated and relaxed model (33) for that example. The lower bound is the initial bound that *fs* obtained by solving model (33) over the entire feasible space. The inital gap represents the percentage difference between the initial lower and upper bounds. When there is a zero initial gap it means that the first relaxed solution is feasible in the original problem thereby corresponding to the global solution. The column for nodes gives the total number of subproblems that where solved before converging to the global solution. A relative tolerance of 0.01 was used, except for example 2 were exact convergence was obtained after branching and example 12 for which a tolerance of 0.02 was used. It is important to note that the initial lower bound is tight and that it corresponds to a good estimation of the global solution. The largest differences are for example 1 with a 25% of difference and for example 12 with a 7% difference. The LP time refers to the time used to solve each relaxed model and the NLP time is the time used for solving a nonconvex model. It is possible to do updates using the previous LP solution and in this form have a more efficient implementation. The times are in seconds and the problems were solved on an IBM RS600/530 using GAMS 2.25 (Brooke et al. (1988)). MINOS 5.2 was used to solve the LP problems and CONOPT for the nonconvex NLP problems. A brief description of the example problems 6 to 12 is given below. It includes the specific data for the problem, the objective function and the topology of the network that is the global solution.

Example 6

This example corresponds to example 2 from Floudas (1987). In this case a linear objective function with the same cost coefficients is used and it is given by,

$$\Phi = 2.5\, S_1 + 3.0\, S_2 + 1.5\, S_3 \tag{51}$$

The data for the composition of the products is given in Table 2.

The initial lower bound is Φ_L = 54.25 and an upper bound of 0 = 55.5 is obtained by solving the nonconvex problem. A partition of the feasible region is performed using the composition of component D in the bottom stream of separator 1. The first subproblem (x £ 0.166) yields a lower bound of Φ_L = 55.45 and the second subproblem (x ≤ 0.166) has a solution of Φ_L = 55.8. The latter is greater than the upper bound iand the former is less than 1% of the global solution (see Fig. 17).

Exajnple7

This example is taken from Floudas (1987). The data for this problem is given in Table 3 and the linear objective function is given by:

$$\Phi = 2.5S_1+3.0S_2+L2S3 \quad (52)$$

The initial lower bound is Φ_L = 32.7 and it provides a feasible solution to the nonconvex problem. In this form the global solution (see Fig. 18) is obtained in one iteration. It is interesting to see that this solution also provides a better objective function for the concave objective function used by Floudas (1987) ($ = 10.65 versus $ = 13.68 which is 28% higher)

Example 8

Tills four component problem is taken from Wehe and Westerberg (1987). The data for the products is given in Table 4 and the objective function has the following form:

$$\Phi = 5.0 + 0.5\ Si + 4.0 + 0.3\ S2 + 6.0 + 0.7\ S_3 \quad (53)$$

The first relaxed subproblem has a solution of 0_L =26.76 and it is infeasible for the nonconvex problem. A nonconvex problem is solved using CONOPT with the LP solution as the initial point. An upper bound of 4> = 26.79 Is obtained corresponding to the global solution (see Fig. 19) within a 0.1%.

Example 9

This example corresponds to example 1 from Wehe and Westerberg (1987). Table 5 provides the data for the product flows and the objective function is given by:

$$\Phi = 5.0 + 0.5\ Si + 9.0 + 1.0\ S2 + 3.0 + 0.4\ S_3 + 6.0 + 0.6\ S_4 \quad (54)$$

A initial lower bound of Φ_L = 85.16 is obtained and the upper bound Is $ « 85.65. The difference is 0,5% and the global solution (see Fig. 20) is obtained in one iteration.

Example 10

This problem is taken from Floudas (1987) and the data is given in Table 6. The objective function is given by,

4 « 1.2 Sj + 3.0 Sj + 2.5 S_3 + 1.5 S_4 **(55)**

The JrHtui lower bound is <^ = 156.56 and the upper bound is <Þ = 179.08. After 5 nodes the global solution of $ = 159.48 (see Fig. 21) is obtained.

Example 11

The data for this 6 component problem are given in Table 7 and the objective function has the following form:

4 »i.5 S, + 3.0 Sj + 2.0 S3 + I.OS4 + 4.0 S_5 . **(56)**

The initial lower bound is 0_L = 173 and the upper bound is 4 » 179.11. After five nodes the global solution is obtained (see Fig. 22).

Example 12

This is a 6 component 4 products problem and the data are given in Table 8. The objective function is:

$ *m* 5.0 Si + 3.0 S2 + 2.0 S_3 + 2.5 S_4 + 4.0 S_5 **(57)**

The initial lower bound is $<f_L$ = 362 and the initial upper bound is 0 = 415.6. The global solution of 4> = 388 (with a 2% optimality gap) is obtained after 33 nodes (see Fig. 23).

Conclusions

A general procedure for the global optimization of process networks with multicomponent streams has been proposed. The basic idea relies on a relaxed LP model that is obtained through reformulation-linearization techniques that establish a clear relation between the component flow and the composition models for mass balances. The reformulated model combines both of these providing tighter lower bounds than other relaxations proposed in the previous work. The relaxed linear model has been embedded in a branch and bound procedure to obtain the global optimal solution.

As has also been shown, the algorithm can be further specialized to take advantage of the particular structure of sharp separation networks with single feed and mixed products. Here, it is possible to preprocess the problem to reduce the space over which the search is

conducted. The bounds that are necessary for the estimator functions in the relaxed model can be obtained without having to solve any subproblems. Different types of linear approximations that are nonredundant to the relaxed model are included to obtain a tighter lower bound.

Twelve examples for both general process networks and for sharp separation networks have been presented to illustrate the performance of the algorithm. As has been shown, only a small number of nodes are commonly needed in the branch and bound search to identify the global or e-global solution. Moreover, in many cases the initial lower bound is either the exact solution or a very good approximation to the global solution.

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Appendix A. Reformulation-Linearization to obtain the nonlinear constraints in model MF

The nonlinear equations in model MF. that can be expressed either as (8) or (9), can also be generated from model MX. For this purpose take the concentration model MX and consider equation (5),

$$x_j^k = x_j^l \qquad \text{(A.1)}$$

multiply by the valid bound constraint X_j^k £ 0

xfvfm^xf **(A.2)**

Use equation (5) for component J\

*xfxf**xtXf*^{k} **(A.3)**

Multiply by the valid bound constraints $F^k \geq 0$ and $F^1 \geq 0$,

$F^k x,^k F^l X f^4$ = $F*\wedge^l F^k x\wedge^k$ (A.4)

that it is linearized to yield.

$$f_j^k f_j^l = f_j^l f_j^k \qquad \text{(A.5)}$$

which is precisely equation (8) for the splitters in the individual flow model MF.

Consider again equation (5),

$x,^k = \wedge$ IA.6)

multiply by the valid bound constraints $F^k \geq 0$ and $F^1 \geq 0$,

F^xfF^F'xj'F^{1}' IA.7)

that can be linearized to yield.

$$f_j^k F^l = f_j^l F^k \qquad \text{(A.8)}$$

Define the split fraction £ to be,

? » £ (A.9)

Equation (A.8) can then be expressed as

fjt^tff (A. 10)

which corresponds to equation (9).

Hence, the nonlinear equations (8) and (9) are redundant to model MX. Their linear approximations in general are also redundant in the linear reformulated model MR. Consider equation (A. 10), similarly to (15) one of its linear approximations is given by.

$$f_j^l \geq \xi^{lL} f_j^k + \xi^l f_j^{kL} - \xi^{lL} f_j^{kL} \qquad \text{(A.11)}$$

If there are no particular restrictions in the splitters, then the bounds for the split fraction variable are 0 £ £^{f} ≤ 1 and using them in (A. 11) yields.

$$f_j^1 \geq f_j^{kL} \xi^1 \qquad (A.12)$$

The bound for the individual component flow is given by $f_j^{kL} = x_j^{kL} F^{kL}$; also $x_j^k = x_j^1$ and $\xi^f = \frac{F^1}{F^f}$ which leads to,

$$f_j^1 \geq x_j^{kL} F^1 \frac{F^{kL}}{F^k} \qquad (A.13)$$

The estimator (15) for the same conditions ($F^{1L} \to 0$) is given by

$$f_j^1 \geq x_j^{kL} F^1 \qquad (A.14)$$

Since the factor $\frac{F^{kL}}{F^k}$ is always less or equal than 1. equation (A. 13) is redundant. A similar analisis can be performed for the other estimators. Only when more specific bounds over the split fractions or the individual component flows are known, will these additional estimators be non redundant.

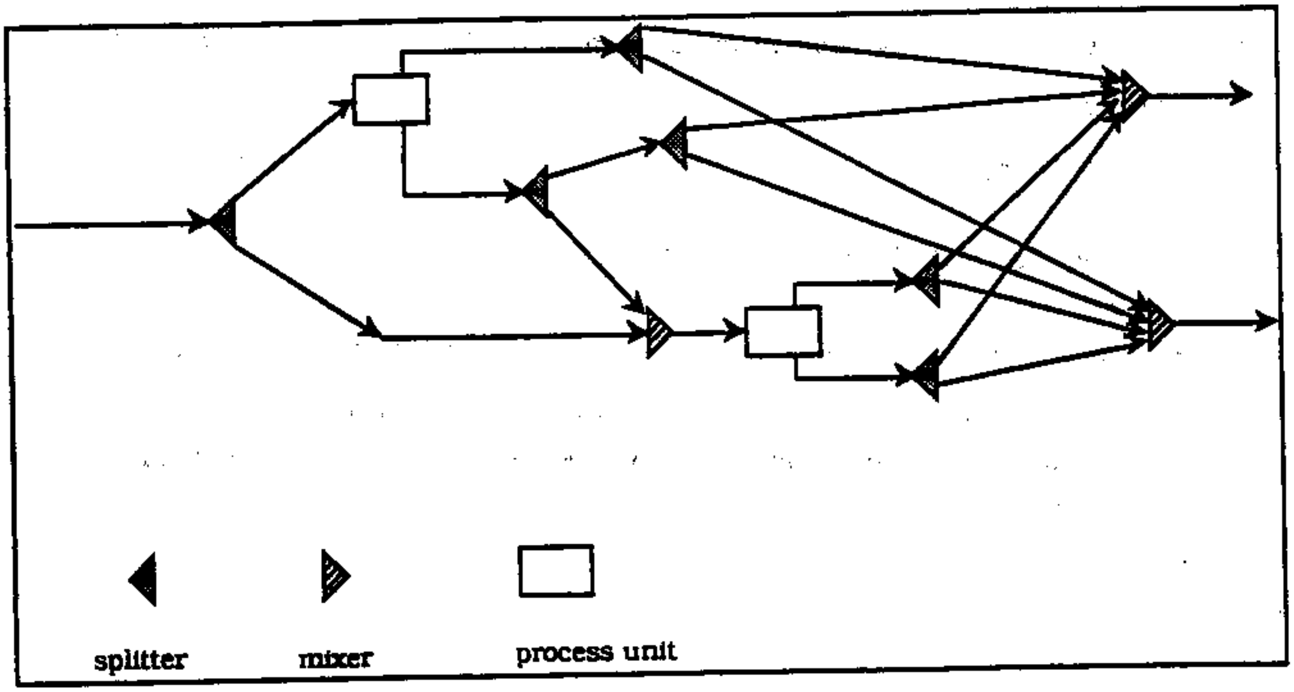


Figure 1. Process network with units, splitters and mixers.

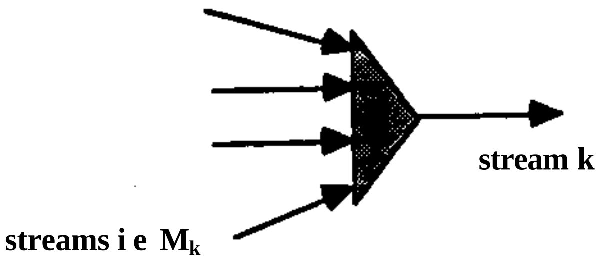


Figure 2. Mixer module.

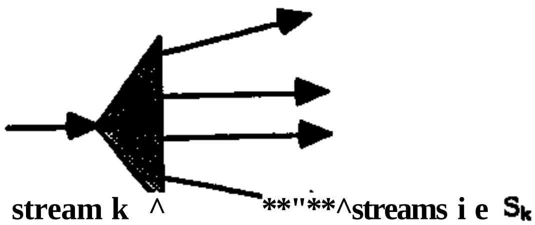


Figure 3. Splitter module.

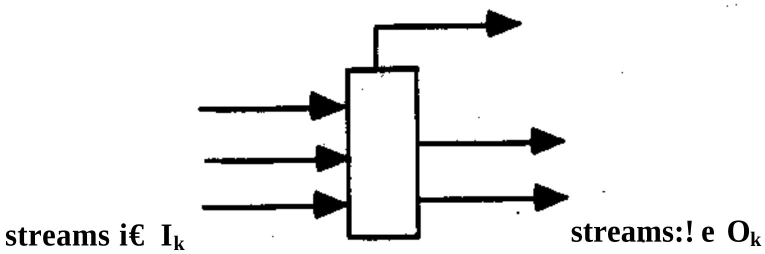
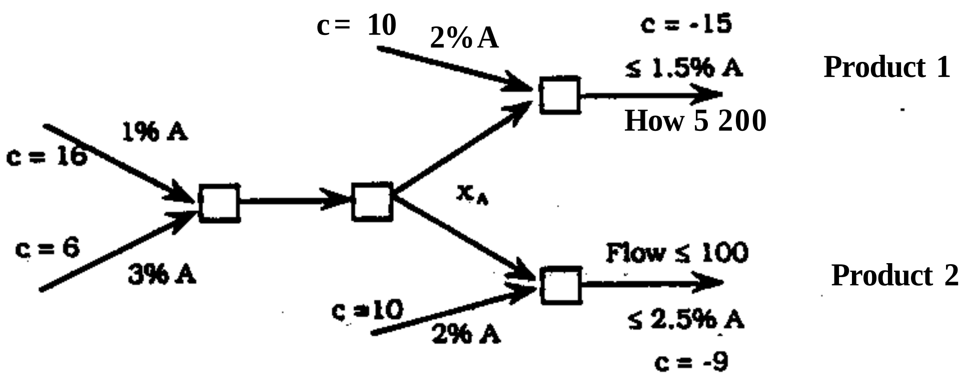
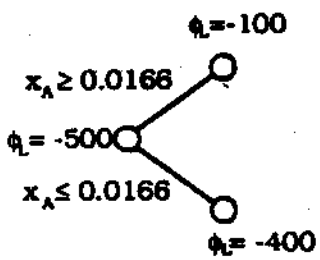


Figure 4. Process unit module.



(a) Network



(b) Tree search

Figure 5. Network and branch and bound search for example 1

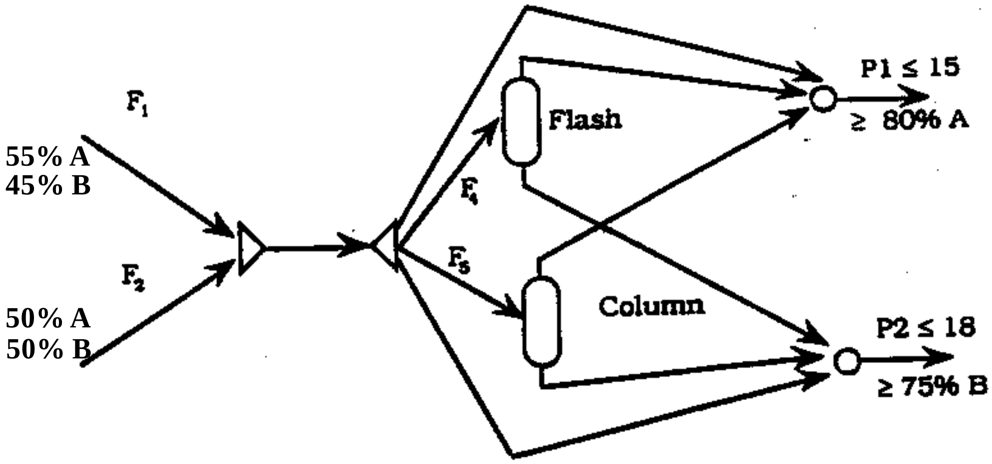


Figure 6. Network for example 2.

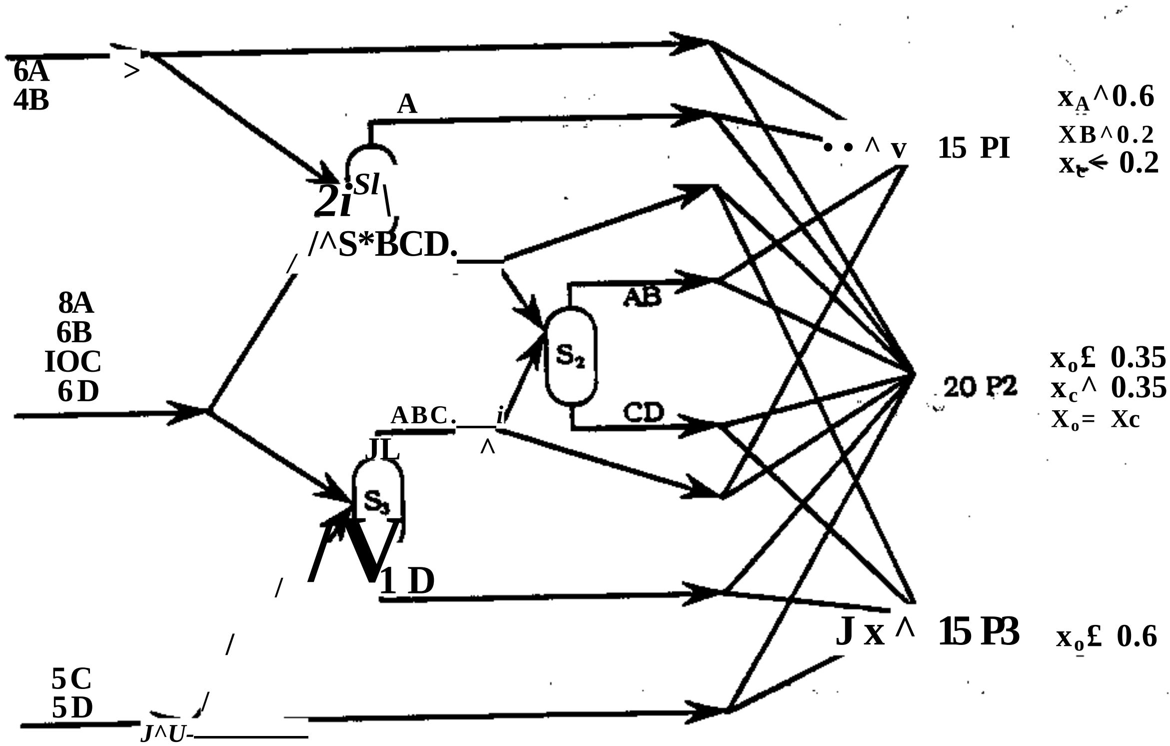


Figure 7. Network for example 3.

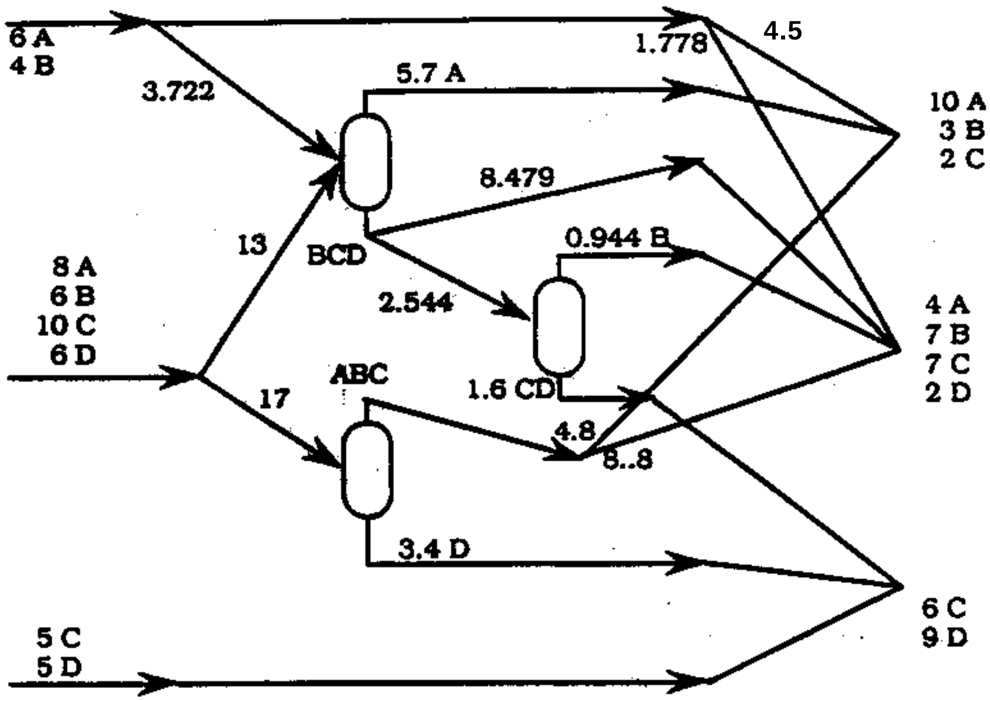


Figure 8. Optimal network for example 3.

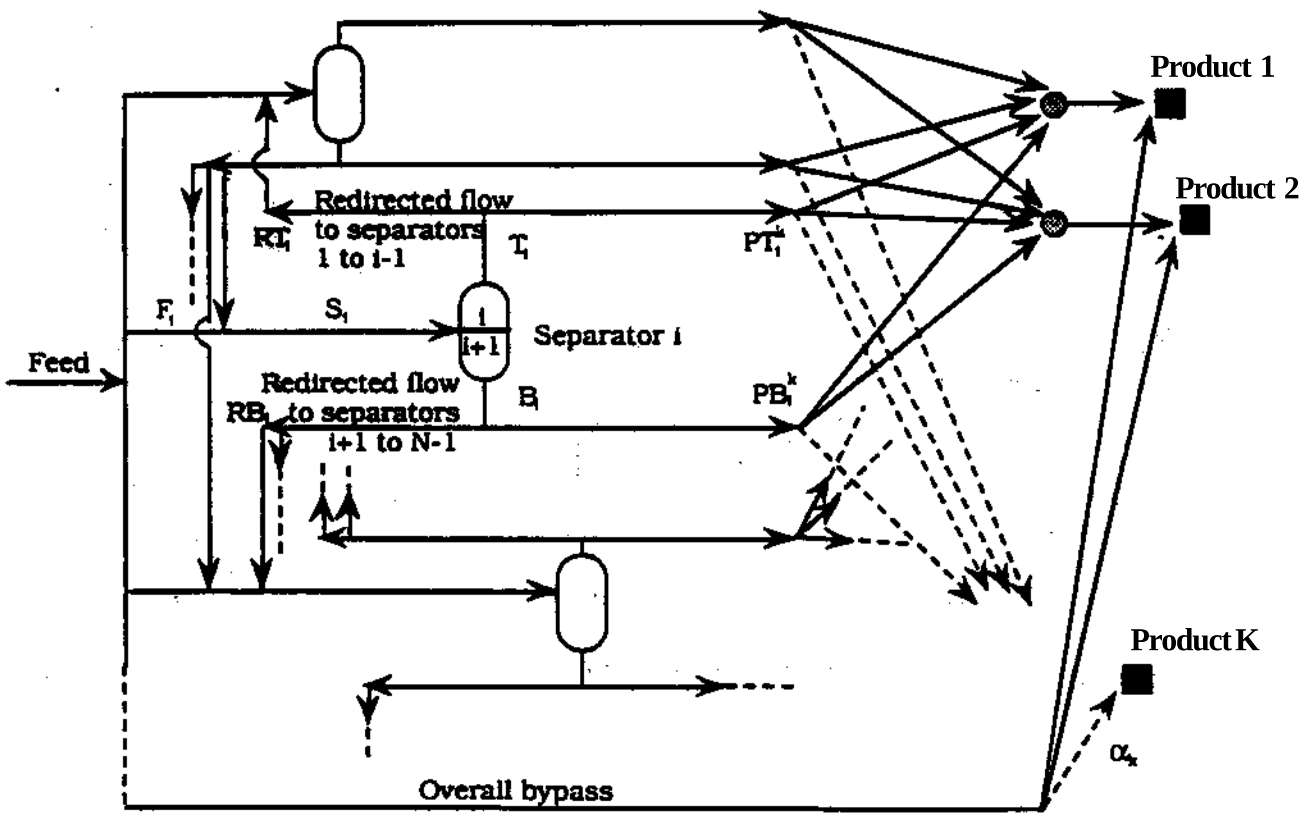


Figure 9. Superstructure for separation with sharp splits and mixed products.

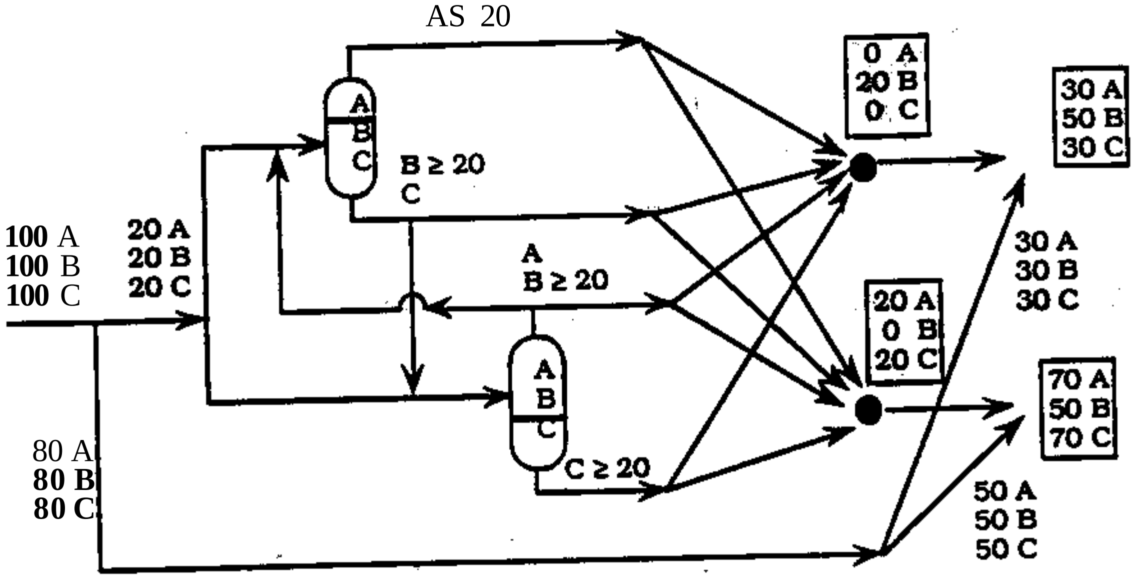


Figure 14. Residual products and key component bounds in example 4.

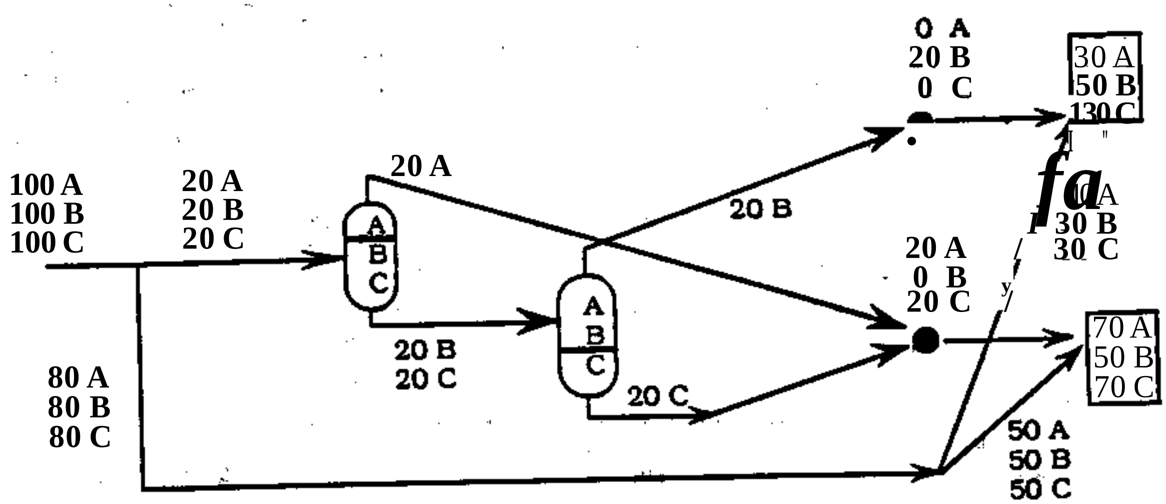


Figure 15- **Global** optimum solution of example 4.

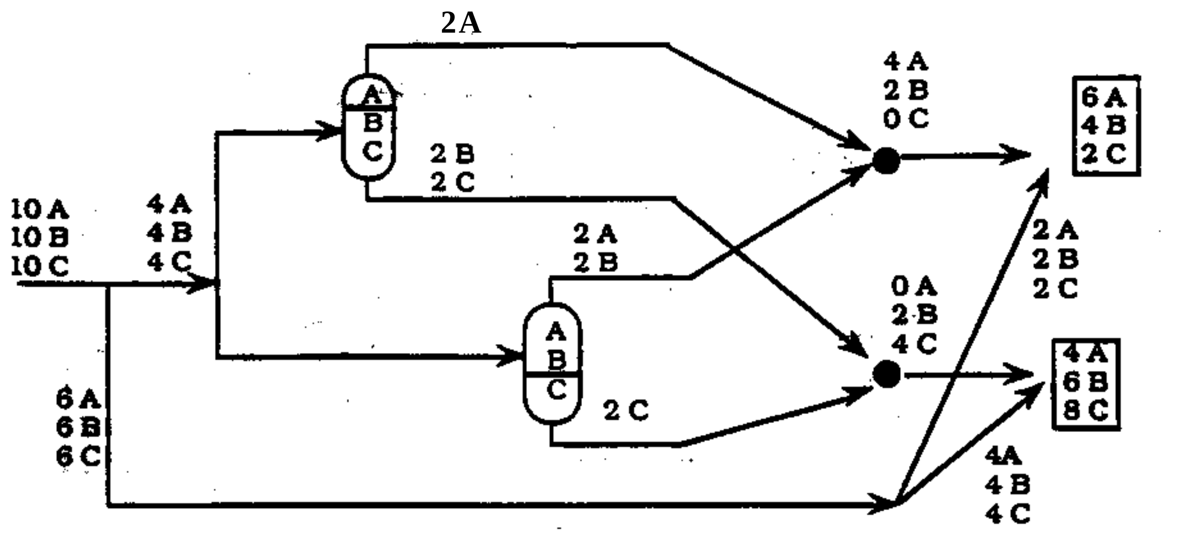


Figure 16. Global optimum solution of example 5.

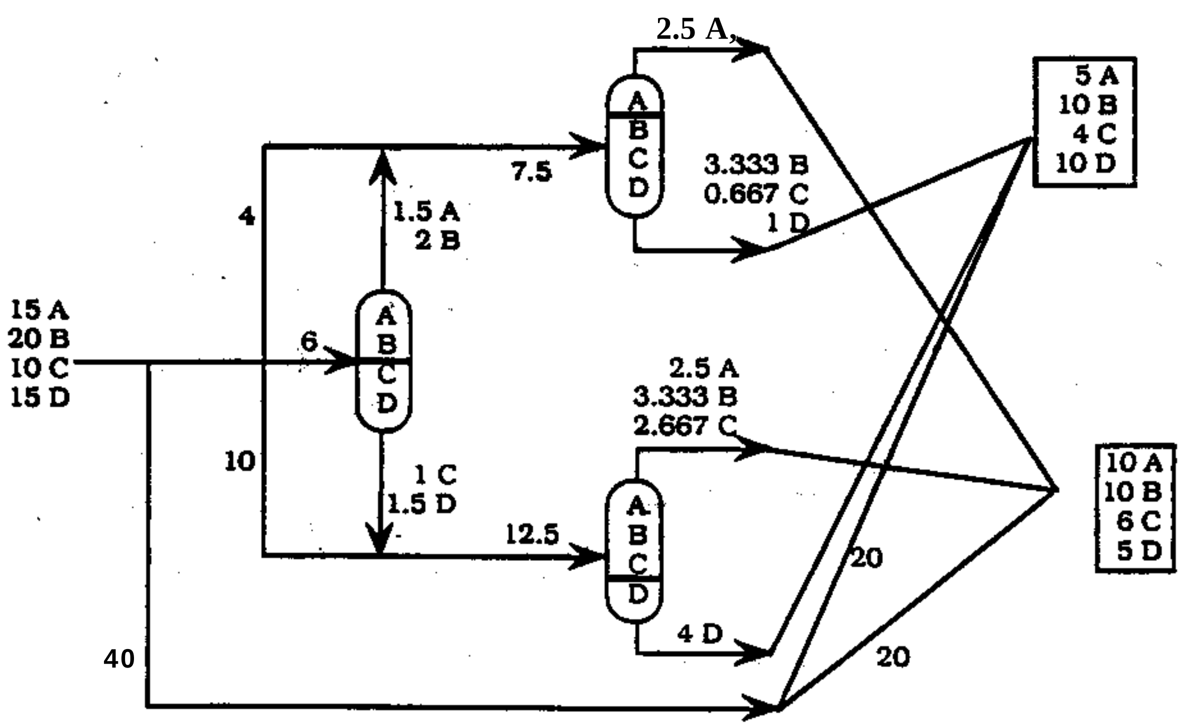


Figure 17. Solution of example 6.

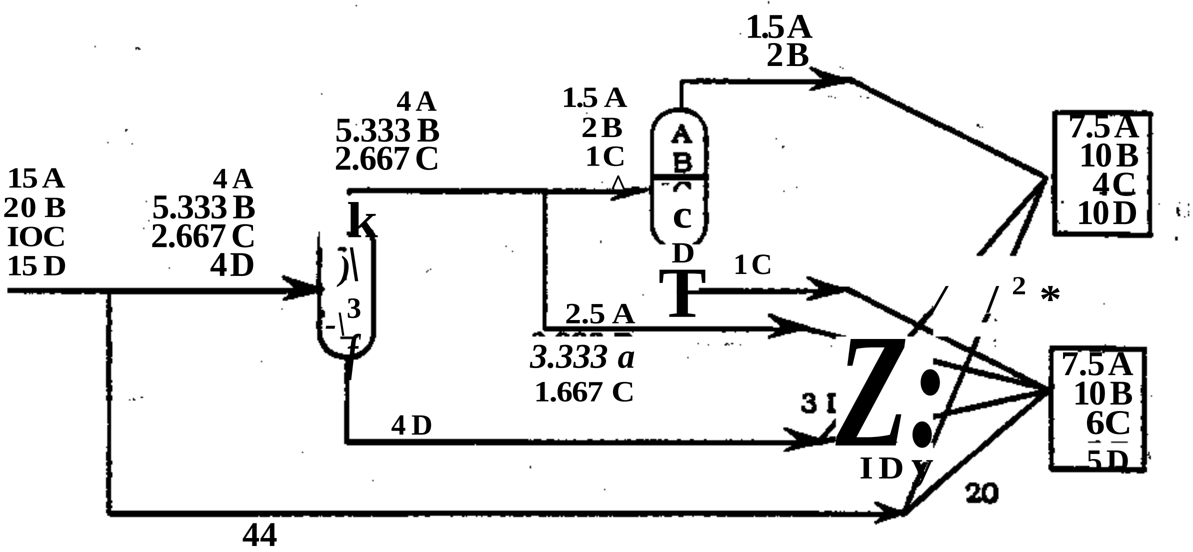


Figure 18. Solution of example 7.

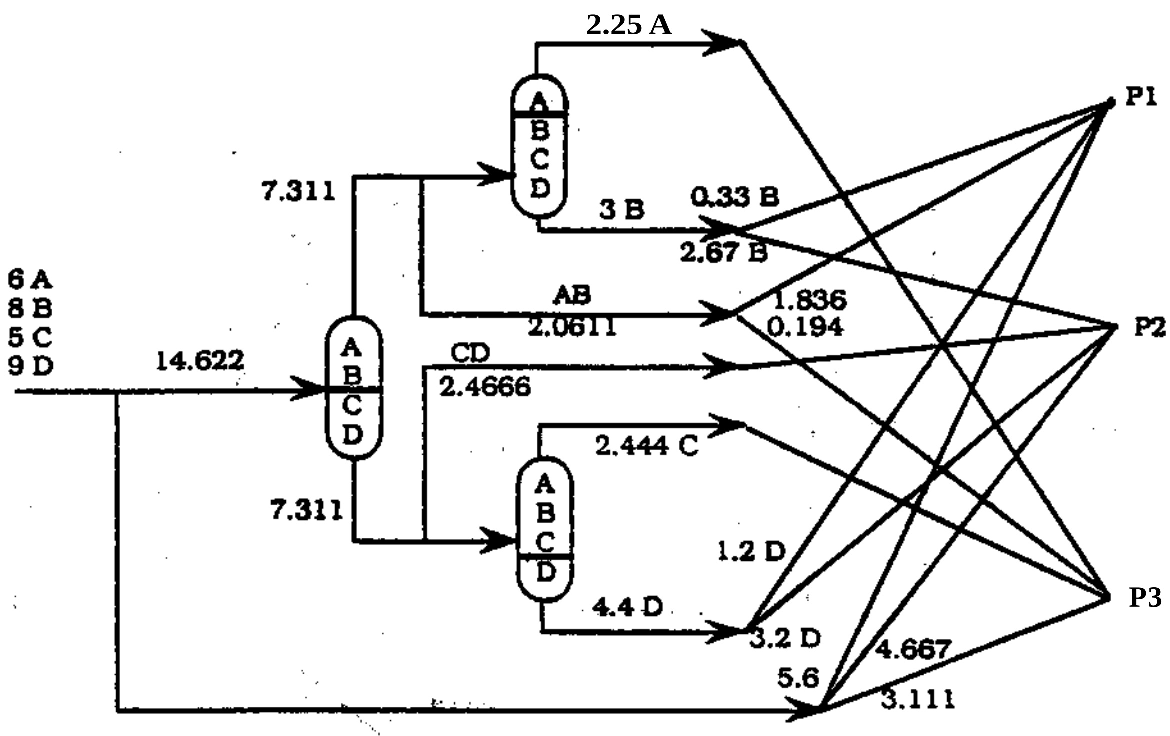


Figure 19. Solution of example 8.

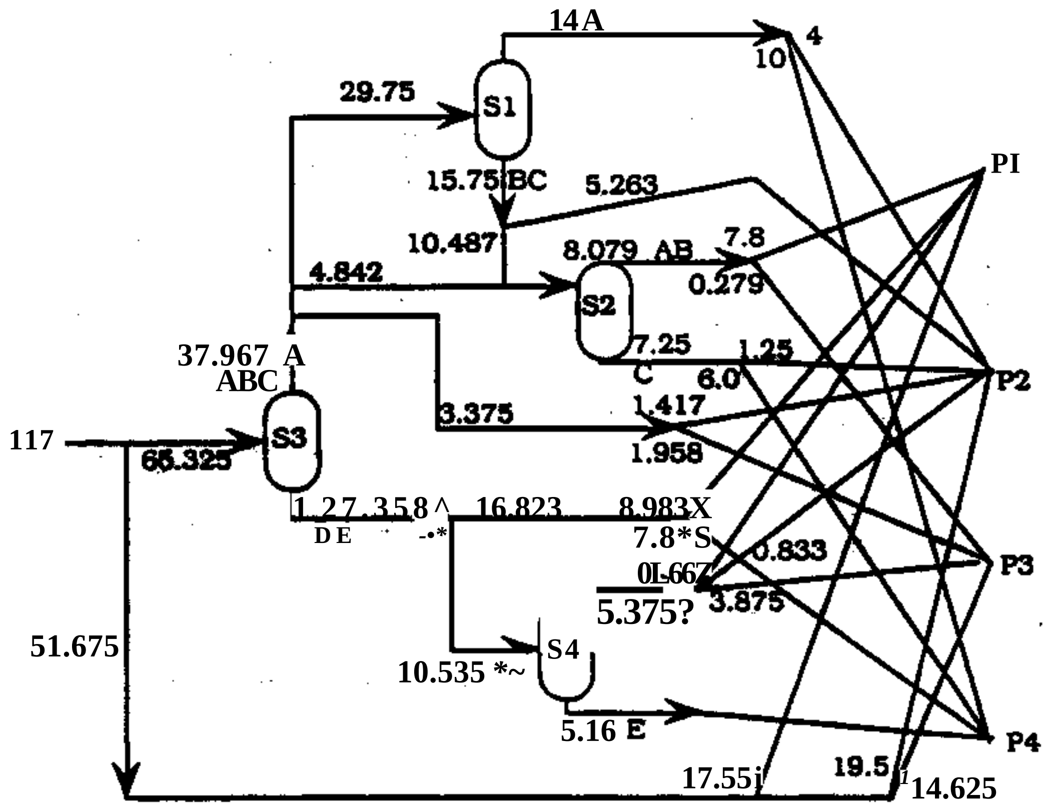


Figure 20. Solution of example 9.

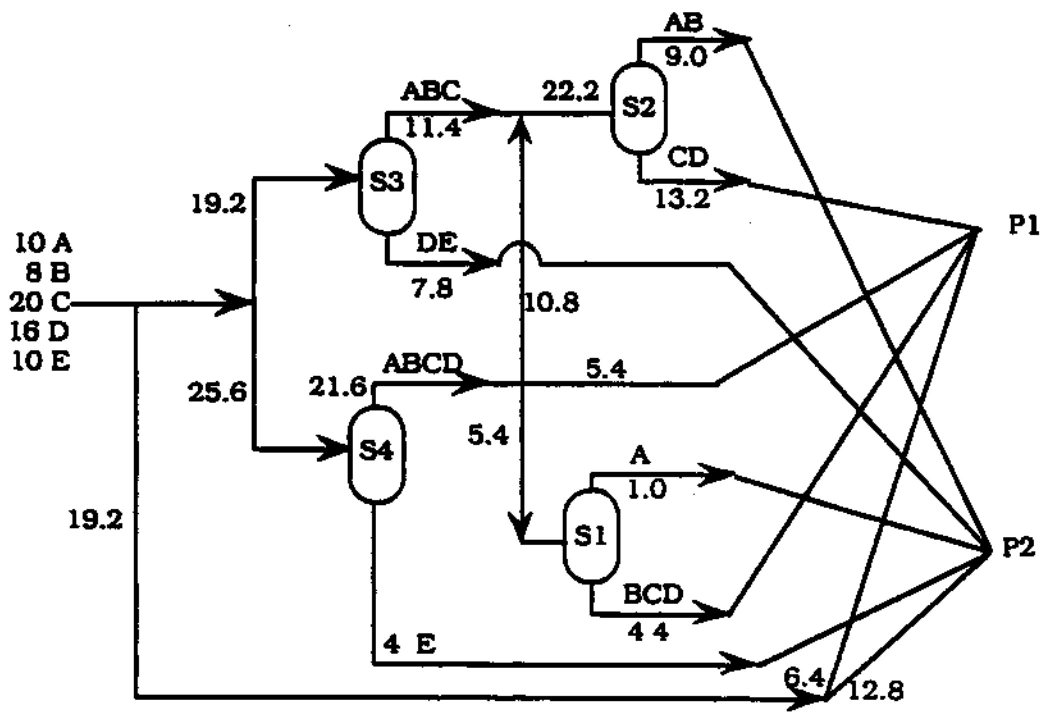


Figure 21. Solution of example 10.

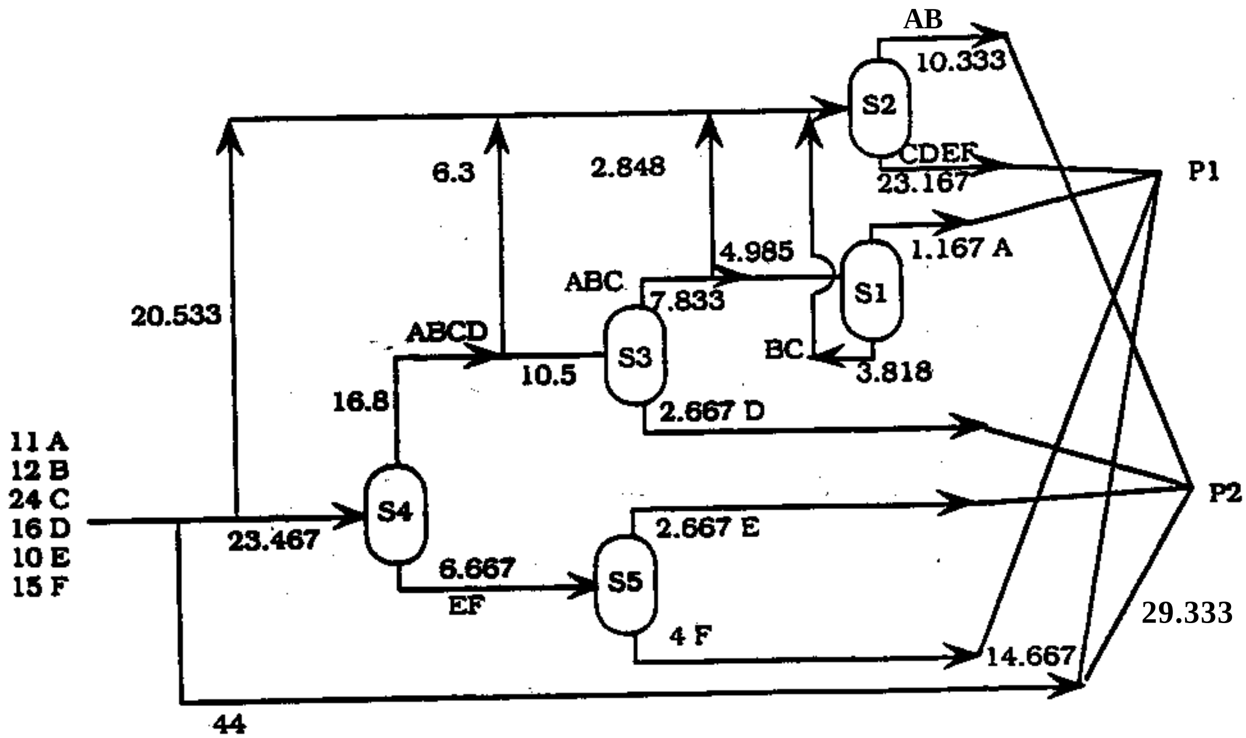


Figure 22. Solution of example 11.

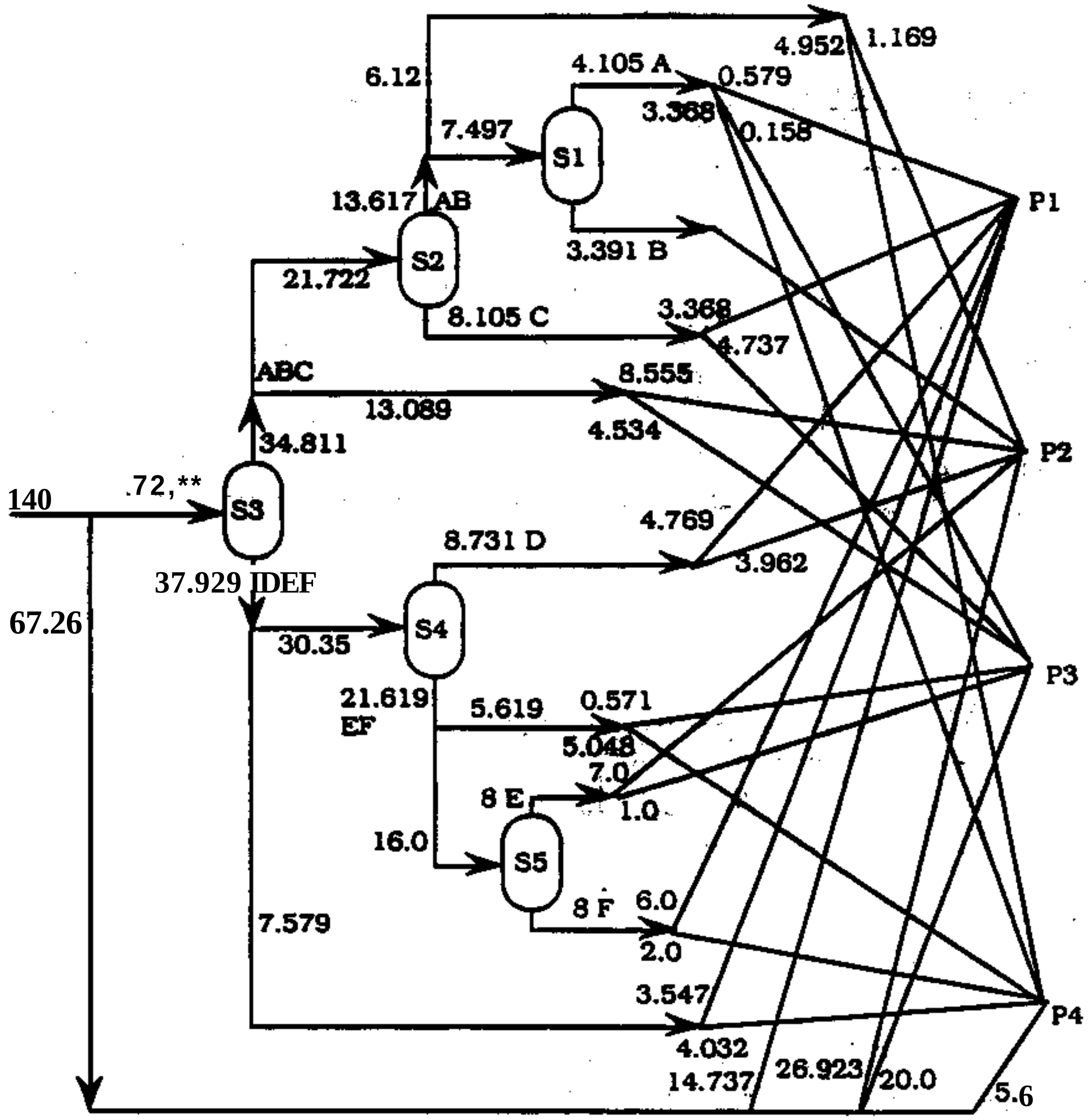


Figure 23. Solution of example 12.

Table 1. Computational results.

	Comp.	WocL	Var.	Lower bound	Initial gap	Global solution	Nodes	LP time	NLP time
Example 1	—	...	29	-500	20'	-400	3	0.05	0.1
Example 2	—	...	35	-513.22	0.3	-511.87	3	0.26	0.3
Example 3			113	138.18	0.4	138.7	1	0.34	0.4
Example 4	3	2	65	1.8639	0.0	1.8639	1	0.13	-.
Example 5	3	2	65	16	0.0	16	1	0.13	..
Example 6	4	2	107	54.25	2.3	55.5	3	0.97	0.4
Example 7	4	2	107	32.7	0.0	32.7	1	0.17	..
Example 8	4	3	125	26.76	0.1	26.79	1	0.23	0.3
Example 9	5	4	281	85.16	0.5	85.65	1	3.08	2.8
Example 10	5	2	225	156.56	12.4	159.48	5	2.59	2.3
Example 11	6	2	350	173	3.5	179.11	5	9.98	8.8
Example 12	6	4	430	362	14.8	388	33	19.8	13.2

Table 2. Data for example 6.

Component	A	B	C	D"	Total
Product 1	5	10	4	10	29
Product 2	10	10	6	5	31
Feed	15	20	10	15	60

Table 3. Data for example 7.

Component	A	B	C	D	Total
Product 1	7.5	10		10	31.5
Product 2	7.5	10	6	5	28.5
Feed	15	20	10	15	60

Table 4. Data for example 8.

Component	A	B	C	D	Total
Product 1	2	3	1	3	9
Product 2	1	4	1	5	11
Product 3	3	1	3	1	8
Feed	6	8	5	9	28

Table 5. Data for example 9.

Component	A	B	C	D	E	Total
Product 1	7	8	3	9	8	35
Product 2	10	3	5	5	4	27
Product 3	5	5	6	7	3	26
Product 4	10	0	6	4	9	29
Feed	32	16	20	25	24	117

Table 6, Data for example 10,

Component	A	B	C	D	E	Total
Product 1	2	2.4	16	8	1	29.4
Product 2	8	5.6	4	8	9	34.6
Feed	10	8	20	16	10	64

Table 7. Data for example 11.

Component	A	B	C	D	E	F	Total
Product 1	3	2	16	8	4	10	4$
Product 2	8	10	8	8	6	5	4$
Feed	11	12	24	16	10	15	88

Table 8. Data for example 12.

Component	A	B	C	D	E	F	Total
Product 1	3	2	6	8	4	10	33
Product 2	8	10	8	8	6	5	45
Product 3	5	4	10	3	11	4	37
Product 4	7	3	1	2	5	7	25
Feed	23	19	25	21	26	26	140

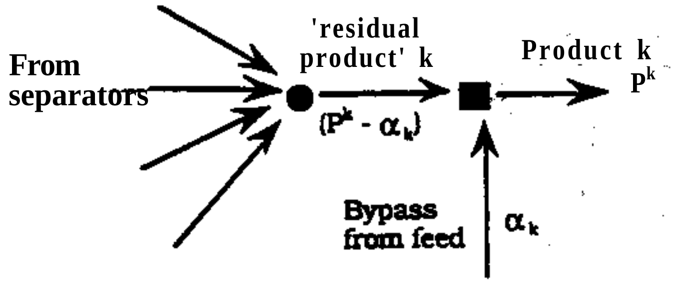


Figure 10. Definition of'residual product'.

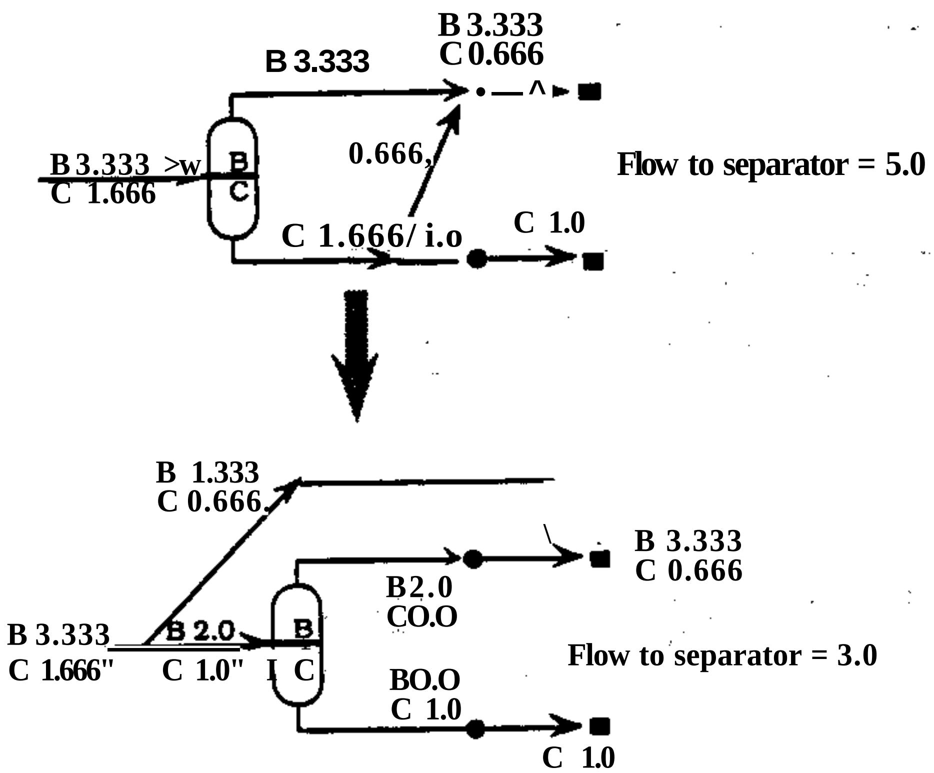


Figure 11. Example of solution without and with a zero component flow in 'residual product".

Global Optimization of Process Networks with Multicomponent Flows

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EDRC 06-164-94

Global Optimization of Process Networks with Multicomponent Flows

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Abstract

This paper deals with the global optimization of networks consisting of splitters, mixers and linear process units and which involve multicomponent streams. Examples include pooling and blending systems and sharp separation networks. A reformulation-linearization technique is first applied to concentration and flow based models in order to obtain a relaxed LP formulation that provides a valid lower bound to the global optimum. This formulation is then used within a spatial branch and bound search. The application of this method is considered in detail for sharp separation systems with single feed and mixed products. Numerical results are presented on twelve test problems to show that only few nodes are commonly required in the branch and bound search.

Introduction

A common source of nonconvexities in the synthesis and design of processes, as well as in flowsheet optimization, are the material flow equations for multicomponent streams. These nonconvex equations involve bilinear terms and they arise in the mass balance equations when the compositions are unknown. There are different equivalent formulations for this type of networks. One alternative is to formulate the mass balance equations in terms of component concentrations* In this form bilinear terms are present in tht equations for the mixer units and the different process units (e.g. sharp separators). A second alternative is to express the mass balances in terms of flows of individual components. This option has the advantage that it involves a smaller number of nonlinear equations. However, the modelling of the splitter units involves bilinear terms that arise due to the condition that the proportions of flows between components have to be the same for the different streams.

The difficulty with the nonconvexities noted above is that they may give rise to optimization problems involving several local optima and numerical singularities that may produce failure in the NLP algorithms. Recently there have been important efforts in the area of global optimization. Examples of algorithms are the ones proposed by McConnick (1976), Floudas and Viswewaran (1990) and Sherali and Alameddine (1992) which can be used to solve bilinear programming problems like the ones that arise in networks with multicomponent streams. For a recent review in the area of bilinear programming see Al-Khayyal (1992).

As for previous work in the design and synthesis of multicomponent process networks Mahalec and Motard (1977) and Nath (1977) developed evolutionary techniques that are based on heuristics to generate a network configuration. Floudas (1987) addressed the synthesis of separation networks with mixed products in which only sharp separators are considered. A superstructure of the process network was proposed and modelled in terms of concentrations. The resulting model is nonconvex and solved with a standard NLP algorithm with no guarantee of global optimality. Floudas and Aggaiwal (1990) solved small pooling and blending problems and sharp separation networks problems using a strategy based on Senders decomposition. In this approach only convex subproblems are solved but there is no guarantee of obtaining the global optimum. Kocis and Grossmann (1989) modelled process networks with multicomponent streams in terms of the individual component flows. They included a set of bounding constraints with binary variables to approximate the nonconvexities that are present in splitters with multiple outputs. Wehe and Westerberg (1987) studied the problem of sharp separation networks with mixed products. They proposed a search procedure that involves the

enumeration of the different separation sequences. The nonconvex equations are dropped and constraints that are valid for each particular sequence with a set of bounds over the key components are included to obtain tighter LP relaxations for each configuration. However, the number of sequences to be examined grows rapidly and there is no guarantee of global optimality*

In some particular cases the nonconvexities in the mass balances can be avoided through the introduction of binary variables. One of these cases is when single choice splitters are present in the flowsheet (Kocis and Grossmann. 1989). Here, it is possible to have a mixed integer ?fryfb;' formulation for the mass balance equations in terms of the individual component flows. Another restricted case for which some nonconvexities can be reformulated is when mining within the network is only allowed for streams of the same concentration. In this form, larger network superestructures must be proposed and the concentrations of the streams are known beforehand. Integer variables are introduced to model the existence of the different streams (e.g. the mixed integer linear formulation for sharp separation networks by Floudas and Anastasiadis, 1988).

The objective of this paper is to present an efficient global optimization method that exploits the particular structure that is present in process networks with multicomponent streams (e.g. pooling and blending systems, sharp separation networks). First a relation is established between formulations based on concentrations and individual flows. This is done following the Reformulation-Linearization technique proposed by Sherali and Alameddine (1992). As will be shown, a linear relaxation Is obtained that is in the space of the concentrations and individual flows which can be used in a branch and bound search is to find the global optimum. Application to the optimal design of sharp separation systems with single feed and mixed products is considered in detail. Different preprocessing techniques that allow tightening of the relaxation problem are presented. The performance of the algorithm is reported on a total of twelve problems.

Modeling with concentrations and individual flows

Consider a process network that consists of splitters, mixers and pfrbcess units tliat are interconnected with multicomponent streams (see Fig. 1). The process iinits that arc considered in this paper are units in which the output flows of the components can be expressed as a linear relation of the inlet flows (e.g. sharp separators, reactor with^known conversion). It is possible to formulate the mathematical model of the process network in terms of the concentrations of the streams, Xj^k. Another possibility is to model the network

using flows of individual components. The former has the advantages that it provides a convenient framework for the evaluation of thermodynamic properties, and in many cases bounds can be expressed in a more natural form. A major disadvantage is that many nonconvex terms (bilinear) are involved in the mass balances for the components. Hie individual component flows formulation is often chosen since it gives rise to a larger number of linear equations and the only nonconvexities are involved in the modelling of the splitters. In these untis it is necessary to enforce that the components maintain the same concentration in each of the streams leaving the splitter. These constraints can be expressed as relations between the different components (Wehe and Westerberg. 1987). One deficiency of this representation is that since many flows can take values of zero, singularities may arise with which conventional nonlinear programming methods may have difficulties to converge. Another alternative is to introduce additional variables that represent split fractions (Kocis and Grossmann. 1989). This involves a larger number of constraints but tends to yield a formulation that numerically is better behaved.

Following are the equations that apply to the mixers, splitters and units using the two alternative representations:

Mixer

A mixer k consists of a set of inlet streams, M_k, and an outlet stream k (see Fig.2).

a) Concentrations

The total mass balance for a mixer k is given by:

$$F^k = \sum_{i \in M_k} F^i \qquad (1)$$

where F^i Is the total flow in stream i. The mass balance for each component j is given by the nonlinear equations,

$$F^k x_j^k = \sum_{i \in M_k} F^i x_j^i \qquad \text{for all } j \qquad (2)$$

where x_j^i is the concentration of component j in the stream i

b) Individual flows

Here it is only necessary to write a mass balance for each component j, given by the linear equations:

$$f_j^k = \sum_{i \in M_k} f_j^i \qquad \text{for all } j \qquad (3)$$

where f_j^i is the flow of component j in stream i.

Splitter

A splitter k has an inlet stream k and a set of outlet streams S^k (see Fig. 3).

cQ Concentrations

The equations for a splitter in terms of the concentrations are given: by the following linear equations

$$F^i = F^k \qquad i \in S_k \qquad (4)$$

$$x_j^i = x_j^k$$ for alii 6 $S_k$ai*dj (5)

$$\sum_j x_j^k = 1 \qquad (6)$$

b) IndividuuaLJlows

The mass balance for each component J is given by

$$\sum_{i€Sfc} f_j^i \text{ sff}$$ forallj (7)

Here, it is also necessary to enforce the condition that the streams leaving the splitter have the same proportions in flow for each component. These relations between components, which are nonlinear, can be expressed in terms of the inlet stream k and a given component J*

fj* tf = fj^k fjV for alii 6 $I_k$andj*j' (8)

A different approach consists of introducing as additional variables the split ratios ^, that represent the part of the inlet flow that goes to the outlet stream i. The nonlinear equations are given by

$$f_j^i = \xi^i f_j^k$$ (braliie $I_k$andj (9)

with $0 \le \xi^i \le 1$.

Process units

In this paper it will be assumed that the outlet streams, i € O_k, in the process units can be expressed as linear relations of the inlet streams, i e I_k (see Fig. 4). This is for instance the case of sharp split separators, separations in which the recovery level is known, or reactors that have a fixed conversion.

a) Concentrations

T

The overall mass balance for process unit k is given by,

IF*= £F (10)

if Ik *«°k

The mass balance for each component] is given by the nonlinear equations:

£ P^F*'^'sPx' forallie $O_k$andj (11)

»*«lk

where p_{jr}^{lk} is a constant for process unit k that gives the distribution of component j in the stream i e O_k coming from streams i^1 € I_k. For a separator unit it is required tliat $_{i \in O_k}$ £ f^* = 1 and £ Pji^{tk} s i . A sharp split separator is one for which II_kl = 1 and IO_kl =2 (top and

$V € I_k$

bottom streams) and for all the components the constant p_{jt}^{ilc} are either 0 or 1.

ffi Uvdiiikixialjlows

Only the **mas** balance for each component is necessary and it is given by:

$$\beta_{jr}^{ik} f_j^{r} \qquad \text{forallie } O_k \text{andj} \qquad (12)$$

A model in terms of individual flows MF consists of the linear equations (3), (7) and (12) plus the nonlinear equations (8) or (9). The model in terms of the concentrations. MX includes the linear equations (1). (4), (5). (6) and (10) plus the nonlinear equations (2) and (11).

Refor"? TI totton and Linearization

In order to avoid the direct use of the nonconvex models MX and MF, there is a relation that can be established between them using the reformulation and linearization technique for bilinear programming models proposed by Shcrali and Alameddine (1992). This technique can be applied to the model MX. First, consider the bounds over the variables present in the bilinear terms (total flow, F^1 and concentrations X_j^1)

$$F^{iL} \leq F^i \leq F^{iu} \qquad (13)$$

$$x_j^{iL} \leq x_j^i \leq x_j^{iu} \qquad (14)$$

Using the bounds in (13). (14) the following constraints can be generated for the bilinear terms in (2) and (UK

$$F^i x_j^i \geq F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

$$F^i x_j^i \geq F^{iu} x_j^i + x_j^{iu} F^i - F^{iu} x_j^{iu} \qquad (16)$$

$$F^1 X_j^4 \text{ £ } F^{*u} x_j^4 + x_j^{iL} F^* - F^{*u} X_j^{IL} \qquad (17)$$

$$F^1 x_j^i \leq F^{iL} x_j^i + X_j^{4u} F^1 - F^{1L} X_j^{*u} \qquad (18)$$

In fact. McCormick (1976) has shown that the constraints in (15M 18) correspond to the convex and concave envelopes of the bilinear terms over the given bounds. The formulation is linearized by the definition of the following variables:

$$\$*¥**\{\bullet \qquad (19)$$

Tfte resultihg model which involves equations (1). (3), (4). (5), (6). (10). (12) and the constraints in (15)-{18) is a linear relaxation of the original nonconvex concentration model. MX. in which the nonlinear equations (2) and (11) have been replaced by the linear equations (3) and (12) from the individual flow model. MF. It is possible to generate additional linear constraints that are redundant to the original nonlinear model, MX, but that can be nonredundant in the linear relaxation of the model (Sherali and Alameddine, 1992: Sherali et

al.. 1992). In particular, consider equation (7) that is the linear component mass balance for the sputters in model MF. This linear equation is not present in the linear relaxation of the concentration model, MX. Take equation (4) and multiply by the valid bound constraint xf £ 0 to get

$$\sum_{i \in S_k} F x_j \wedge F * \wedge \qquad (20)$$

Using equation (5) yields.

$$\sum_{i \in S_k} P x_j \wedge F \wedge \qquad (21)$$

that can be linearized to,

$$\sum_{i \in S_k} f_j^i \text{ "f,k.} \qquad (22)$$

yielding equation (7). Hence, the linear equation for the splitter is valid and it is Included. Hie nonlinear equations (8) or (9) can also be generated in a similar fashion but their linearizations are in general redundant (see Appendix A). They are only useful when the formulation df the problem provides non-trivial bounds over certain components in the outlet streams of a splitter, or when there exist some restrictions over the split ratios for the outlet streams.

Also, the constraints that relate the total flow and the individual flows of a stream can be generated for the splitters. Taking equation (6) and multiply by F^1 yields.

$$F^t \sum_j x_j^k = P \qquad (23)$$

Using the constraints $xf = x_j^k$ in equation (23) and linearizing with $f_j^1 = F^1 xf$ yields.

$$F\text{-}\sum_j f \qquad (24)$$

Based on the above it is possible to obtain a reformulated model MR that involves concentrations, total flows and component flows, and which bounds the solution of the original problem. The following equations are given for model MR:

a) Objective function. 0. which is expressed in terms of individual or total flows,

b) Mixer equations, which are expressed ta terms of the total and individual component flows.

$$F^k = \sum_{i \in M_k} F^* \qquad (1)$$

$$f_j^k = \sum_{t \in M_k} f_j^\wedge \qquad \text{for all } j \qquad (3)$$

c) Splitter equations, that are expressed in terms df the individual component flows and the concentrations of the streams

$$\sum_{i \in S_k} F^i = F^k \qquad (4)$$

$$x_j^i = x_j^k \qquad \text{for all } i \in S_k \text{ and } J \qquad (5)$$

$$\sum_J x_j^k = 1 \qquad (6)$$

$$\sum_{i \in S_k} f_j^i = f_j^k \qquad \text{for all } J \qquad (7)$$

d) Process units equations, that are given in terms of the total and individual component flows

$$\sum_{i \in ifc} F^i = \sum_{i \in Ofc} F^i \qquad (10)$$

$$f_j^i = \sum_{r \in I_k} \beta_{jr}^{ik} f_j^r \qquad \text{for all } i \in O_k \text{ and } j \qquad (12)$$

e) Relation between the total flow and the individual component flows

$$F^i = \sum_J f_j^i \qquad \text{for all streams} \qquad (24)$$

f) Linear estimators, relate the individual component flows with the total flow and concentrations.

$$f_j^i \geq F^{iL} x_j^i + x_j^{iL} F^i - F^{iL} x_j^{iL} \qquad (15)$$

fji S F ^ + x ^ P - F 1 " ^ " for alli€ S_k (16)

fji ≤ jiu ^i ^ x^tL pi. pu x^iL i s k € splitters t!7)

$$f_j^i \leq F^{iL} x_j^i + x_j^{iu} F^i - F^{iL} x_j^{iu} \qquad \text{for all } j \in J \qquad (18)$$

fj Bounds on flows and concentrations

$$F^{iL} \leq F^i \leq F^{iu} \qquad (13)$$

$$x_j^{iL} \leq x_j^i \leq x_j^{iu} \qquad (14)$$

In previous approaches (Wehe and Westerberg, 1987; Kocis and Grossmann, 1989) looser approximations of the nonlinear terms were used. In both cases, the nonconvex problem (MF) was relaxed to a linear model by dropping the nonlinear equations (8) or (9). Equations that approximate the difference relation between the components were considered (Kocis and Grossmann, 1989). They were based on the difference that exists at the inlet of the splitter between the flowrate of the components and required the introduction of binary variables.

Outline of global optimization method

Model MR can be applied to predict lower bounds to the global optimum in the optimization of pooling and blending problems and in the synthesis of separation systems. The reason is that model MR provides a valid relaxation of the original feasible region since the nonlinear equations (2) and (11) in model MX are not considered, and the valid linear equations (3), (7), (12) and (15)-(18) are included. The proposed global optimization algorithm relies on the solution of the relaxed problem MR within a spatial branch and bound enumeration. The outline of the algorithm is as follows (for a more detailed description of step 4 see Quesada and Grossmann (1993))

0. Preprocessing (optional)

Determine bounds on the variables involved in the nonconvex terms, that is total flows. F*. and concentrations, *xf*. Apply any additional preprocessing specific to the structure of the problem in order to further bound or fix variables.

1. Lower Bound

Solve model MR over a given subregion (initial subregion is the complete feasible region) minimizing a convex objective function Φ. If Φ is linear the model is an LP.

2. ***Upper Bound***

Any feasible solution to the nonlinear model provides an upper bound. Heuristic techniques can be employed to obtain good feasible solutions or the original problem. MF. can be solved using the solution of model MR as a good initial point. If the solution of problem MR is feasible it provides an upper bound.

3. ***Convergence***

If the lower bound of a subregion is sufficiently close or above the upper bound discard that subregion. If no subregions are left the global solution corresponds to the best upper bound.

4. ***Branch and Bound***

Partition the remaining subregions into a sell of disjoint subproblems. Repeat steps 1-3 for each of the new subregions.

Remarks

The preprocessing step plays an important role in the above algorithm. It is during this step that initial bounds for the variables involved in nonconvex terms are obtained. The quality of

these bounds affects the tigthness of the lower bound since they are part of the estimator equations (15)418). Additionally, these bounds affect the performance of the algorithm because they define the search space over which the branch and bound procedure may have to be conducted.

In some cases, as described later in this paper, it is possible to exploit the particular structure of the process network and generate bounds for the variables without having |o solve any subproblems. Furthermore, during this preprocessing step additional constraints can be generated for predicting a tighter lower bound of the global optimum can be obtained.

Some of the linear mass balances and the estimator equations are redundant in the nolinear formulations, MF and MX. These equations become nonredundant in the refonnulated model, MR, and for that reason it is important to write a complete formulation of the network. However, this model can present some redundancies that can be easily identified and eliminated to reduce the size of the model. This is the case for the concentration variables used in the splitters. Model MR uses different sets of concentrations variables for the inlet and outlet variables of a split unit. In practice, it is only necessary to define the concentration of the component in the splitter and use the same variables for all the splitter streams. Also, some redundancies can occur with the total flow variables. These ones are necessary for the streams in the splitters but they might be redundant and eliminated in the other untis if they do not appear in other part of the model or in the objective function.

If the solution of model MR is feasible for the original nonconvex problem then it corresponds to the global optimal solution. When the solution to the model MR is not feasible it is necessary to follow a branch and bound procedure to search for the global optimum. This procedure requires a valid upper bound on the global optimum. This can be generated through heuristics or by solving directly the nonconvex model. For this purpose, the process network model is formulated in terms of the individual component flows and the nonconvex equations for the splitters are included. Equation (9J was also used in this work to model the splitters due to it is better numerical behavior. The solution to the model MR was used for the good initial point. In many instances, it was not possible to solve these nonlinear problems with MINOS 5.2. The nonlinear models were solved using CONOPT in GAMS 2.25.

During the branch and bound procedure a tree search is generated. Of the set of open nodes, these are the nodes that have a lower bound that is e-smaller than the current upper bound, the node with the smaller upper bound is selected to branch on. The splitter units are the units that are approximated, and of these, the splitter that has the largest difference

between its approximated and actual individual component flows is selected. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. The branching is done in the selected splitter over the concentration of the component that has the largest difference.

First, the actual concentrations for the individual component flows in the LP solution O for the splitters are calculated by,

$$z_j^k = \frac{f_j^{k*}}{F^{k*}} \qquad \text{for all the inlet streams to splitter} \qquad (26)$$

the splitter unit m is then selected according to the equation.

$$m = \arg\max_{k \in split}\left\{ \sum_{i \in S_k} \sum_{j=1} |f_j^i - z_j^k F^i| \right\} \qquad (27)$$

Equation (27) represents the total difference between the LP solution for the flows after the splitter and the actual value of these flows considering the concentrations before the splitter. Once the splitter has been selected, the component in that sputter that has the largest difference. J\ is selected by.

$$j^f = \text{argmax}_j\left(\sum_{i \in S_m} |f_j^i - z_j^m F^i| \right) \qquad (28)$$

Hie following branching constraints are then used:

$$x_{j'}^m \le z_{j'}^m \text{ and } x_{j'}^m \ge z_{j'}^m \qquad (29)$$

To improve the upper bound it might be necessary to solve additional nonconvex problems. These can be solved after a given number of nodes using the solution of the node with the smallest upper bound as the initial point. In this work if there was no significant change in the lower bound df the new nodes with respect to the lower bound of the parent node (< 1%) a new nonconvex problem was solved.

Example 1

Consider the following pooling and blending problem by Harveley (1978). Two streams that have components A and B are mixed in a initial mixer a then go through a splitter to obtain two streams than can be mixed with an additional stream (see Fig. 5a). Two different products can be obatined and there are constraints on the concentration of component A in these products. The objective function consists of mimizing the cost that is given by the total flow of the streams times the cost coefficients. c_t, given in Fig. 5.

This problem has two local solutions. One has an objective function Φ = -100 and consists of *otify* producing product 2. The other local solution, that corresponds to the global optimum, has an objective of 4> = -400 and here only product 1 is produced.

Model MR is formulated for this problem and the initial lower bound is Φ_L = -500. The nonlinear model, MF, is solved using the solution of model MR as the initial point and an upper bound of • a -400 is obtained* Since there is a gap between the bounds of the global solution a partition is performed- There is only one splitter that needs to be approximated and since there are only two components it is irrelevant which one is selected since the composition bounds are related (eg. x_B^L s i - x^{*u}). The actual value of the composition of A in the solution of model MR is used as the branching point (x^* = 0.0166) to generate two new subproblems. The first subproblem (x_A £ 0.0166) has a lower bound of Φ_L = -100 and the second subproblem ($x^* \leq$ 0.0166) has a solution of fa = -400 (see Fig. 5b). Both of these bounds are greater or equal than the upper bound, therefore the global solution has been found (x_A = 0.01).

Example 2

Tbe next example is a separation problem taken from Kocis and Grossmann (1987). The original problem has binary variables in* the formulation and they have been fixed to 1 for this example (see Fig. 6).

The top stream of the flash unit has 85% of the inlet flow of A and the bottom stream has 80% of the inlet flow of component B. In the column. 97.5% of the inlet flow of A goes to the top whereas 95% of the inlet flow of B goes to the bottom stream. The total flow to the flash unit and the column have to be greater than 2.5 and smaller than 25, whereas the total flow of each of the two feed streams has to be less than 25. The objective function is given by,

$$\Phi \text{ ss } 52 + 10 F! + 8 F_2 + F_4 + 4 F_5 - 35 P,^A - 30 P_2^B \qquad (30)$$

The initial lower bound for this problem is Φ_L = -513.22 and it is infeasible for the original NLP model. A nonconvex problem is solved using CONOPT with the solution of model MR as the initial point obtaining an upper bound of Φ = -511.87 and the relative gap is only 0.3%. Again only one splitter is present in the network and a partition can be performed using the concentration of component A in this splitter. The lower bounds for the new two subproblems are 0L = -511.87 (x_A £ 0.5121) and 0_L = -511.80 ($x_A \geq$ 0.5121). Both solutions are greater or equal than the upper bound and the global solution has been obtained. In the global solution F_x ss 8 and F_2 = 25, and 11% of the inlet flow to the splitter is directed to the flash. 76% to the column and the rest bypassed to P_2.

Example 3

This example corresponds to a separation problem with three feeds and three product streams. The network configuration and product specifications are given in Fig. 7. The objective function is given by

$$\phi = 4\, S_1 + 1.5\, S_2 + 4\, S_3 \qquad (31)$$

The initial lower bound is ϕ_L = 138.18 and the nonconvex problem MP is solved obtaining a solution of ϕ « 138.7. The gap between these bounds is less than 0.4 %. The global optimum for this tolerance is shown In Fig. 8.

Sharp separation networks

In order to illustrate the application of the above algorithm to a specialized case where the structure can be further (exploited, the problem that will be considered is the synthesis of separation networks with single feed and mixed products that consist of sharp separators and bypasses. It is assumed that a single feed with N components must be separated into K specified multicomponent product streams. The components are ordered from the lightest to the heaviest.

A modification of the superstructure proposed by Floudas (1987) for this problem is used (see Fig. (9)). The superstructure consist of N-l separators. Separator 1 performs the task of removing component number 1 to number 1 at the top of the separator and components number i+1 to N at the bottom of the separator. The feed to the network is split into N-l streams, F_t. that go to the separators and K streams, a_k. that bypass the network to go to the products. Each stream F< is mixed before the separator i with streams that come from the top and bottom streams from the other separators to obtain the inlet stream to separator i, St.

The outlet streams of separator i are the top. T_{l9} and the bottom, B_t. These streams, T« and B{, are each split into streams, PT_t^k and PB<^{k} respectively, that go to the K products and into streams, KIV' and RB/\ that are redirected to the other separators. The top stream of separator 1, T_t, can be redirected only to the separators 1 to i-1 since it can only contain components number 1 to i. It would not be optimal to send part of this stream to any separator from i+1 to N since no separation would be achieved and a bypass of these separators would achieve the same separation with smaller (lows. *KTf* is the flow redirected from the top of separator i to separator i\ In the same fashion that with the top stream, the bottom of separator i, B_t, can be redirected only to separators i+1 to N-l since it can only

contain components that are separated by these sharp separators. $RB_i^{i'}$ is the redirected flow from the bottom stream of separator i to separator i'.

Model

It will be assumed that the objective function can be expressed as a linear function that depends on the total flow to each separator. The model expressed in terms of concentrations and total flows has the following form:

$$\min \Phi = \sum_{i=1}^{N-1} c_i S_i \qquad (32.1)$$

$$\text{s.t. } Feed = \sum_{i=1}^{N-1} F_i + \sum_{k=1}^{K} \alpha_k \qquad (32.2)$$

$$f_{ij} = F_i z_j \quad \text{for all i and j} \qquad (32.3)$$

$$S_i = F_i + \sum_{i'=1}^{i-1} RB_{i'}^{i} + \sum_{i'=i+1}^{N-1} RT_{i'}^{i} \quad \text{for all i} \qquad (32.4)$$

$$S_i xs_{ij} = f_{ij} + \sum_{i'=1}^{i-1} RB_{i'}^{i} xb_{i'j} + \sum_{i'=i+1}^{N-1} RT_{i'}^{i} xt_{i'j} \quad \text{for all i and j} \qquad (32.5)$$

$$S_i = T_i + B_i \quad \text{for all i} \qquad (32.6)$$

$$T_i xt_{ij} = S_i xs_{ij} \quad \text{for all i and ord(j)} \leq i \qquad (32.7)$$

$$xt_{ij} = 0 \quad \text{for all i and ord(j)} > i \qquad (32.8)$$

$$B_i xb_{ij} = S_i xs_{ij} \quad \text{for all i and ord(j)} > i \qquad (32.9)$$

$$xb_{ij} = 0 \quad \text{for all i and ord(j)} \leq i \qquad (32.10)$$

$$T_i = \sum_{i'=1}^{i-1} RT_i^{i'} + \sum_{k=1}^{K} PT_i^k \quad \text{for all i} \qquad (32.11)$$

$$B_i = \sum_{i'=i+1}^{N-1} RB_i^{i'} + \sum_{k=1}^{K} PB_i^k \quad \text{for all i} \qquad (32.12)$$

$$P^k = \sum_{i=1}^{N-1} PT_i^k + \sum_{i=1}^{N-1} PB_i^k + \alpha_k \quad \text{for all k} \qquad (32.13)$$

$$p_{kj} = \sum_{i=1}^{N-1} PT_i^k xt_{ij} + \sum_{i=1}^{N-1} PB_i^k xb_{ij} + \alpha_k z_j \quad \text{for all k and j} \qquad (32.14)$$

$$\sum_{ord(j)=1}^{i} xt_{ij} = 1 \quad \text{for all i} \qquad (32.15)$$

$$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1 \quad \text{for all i} \qquad (32.16)$$

$$\sum_{ord(j)=1}^{N} xs_{ij} = 1 \quad \text{for all i} \qquad (32.17)$$

$$S_i, T_i, B_i, F_i, RT_i^{i'}, RB_i^{i'}, PT_i^k, PB_i^k, \alpha_k, f_{ij}, xs_{ij}, xt_{ij}, xb_{ij} \geq 0$$

The parameters Feed, zj, P^k and p_{kJ} represent the total feed, composition of the feed, total flow of product k and component flow of component j in product k. respectively. The variables xsq, xt_{fj} and xb«j are the concentrations of component j in the inlet stream to separator i. top of separator i and bottom of separator i, respectively.

The objective function (32.1) is a linear function of the inlet flow to the separators. Equation (32.2) is the total mass balance in the inlet splitter and equation (32.3) is the component mass balance. Equations (32.4) and (32.5) are the total and component mass balances for the mixer i before the separator L The material balances for separator i are given by equations (32.6). that is the total mass balance for the separator, equations (32.7) and (32.8) that are the component balances for the top stream and indicate that nothing from components number 1+1 to N-l is in the top of the separator, and equations (32.9) and (32.10) that are the component mass balances for the bottom streams. Equations (32.11) and (32.12) are the overall mass balances for the splitters of the top and bottom streams after separator i. The equations that state that the concentrations of the outlet streams should be the same that the inlet stream in a splitter have been already substituted. Finally, equations (32.13) and (32.14) are the overall and component mass balances for the mixer for product k.

Model (32) corresponds to a formulation of the type of model MX where the distribution coefficients are known and restricted to 0 or 1. Some simplifications have been made to avoid including many irrelevant variables (e.g. not to define concentrations for the streams that go the top i to product k). Although, some of the linear constraints in this formulation are redundant, they can become nonredundant in the linear relaxation as will be shown in Example 4.

Equations (32.5), (32.7), (32*9) and (32.14) involve nonconvex terms. This model can be reformulated as in model MR by introducing individual component flows and the linear equations (15)-(18) and (7) according to the approach illustrated eaiiier to obtain a model in the form of model MR. The resulting reformulated model is as follows,

$$\min \ \$' = \sum_{i=1}^{N-1} c_4 S_4 \qquad (33.1)$$

$$\text{st } \ Feed = \sum_{i=1}^{N-1} F_t + \sum^{K} \alpha_k \qquad (33.2)$$

$$f_{ij} = F_i z_j \qquad \text{for all i and j} \qquad (33.3)$$

$$S_i = F_i + \sum_{i'=1}^{t-1} RB_{i'}{}^{i} + \sum_{i'=i+1}^{N-1} RT_{i'}{}^{i} \qquad \text{for all i} \qquad (33.4)$$

$$s_{ij} = f_{ij} + \sum_{i'=1}^{i-1} rb_{i'j}{}^{i} + \sum_{i'=i+1}^{N-1} rt_{i'j}{}^{i} \qquad \text{for all i and j} \qquad (33.5)$$

$S_i = T_i + B_i$	for all i	(33.6)
$t_{ij} = s_{ij}$	for all i and ord(j) £ i	(33.7)
$t_{ij} = 0$	for all 1 and ord(j) > 1	(33.8)
$b_{ij} = s_{ij}$	for all i and ord(j) > i	(33.9)
$t_{ij} = 0$	for all i and ord(j) ≤ i	(33.10)
$T_i = \sum_{i'=1}^{i-1} RT_i^{i'} + \sum_{k=1}^{K} PT_i^k$	for all i	(33.11)
$B_i = \sum_{i'=i+1}^{N-I} RB_i^{i'} + \chi PB_i^k$	for all 1	(33.12)
$P^k = \sum^{N-I} PT_i^k + \sum_{i=1}^{N-I} PB_i^k + \alpha_k$	for all k	(33.13)
$p_{kj} = \sum_{i=1}^{rt-i} pt_{ij}^k + \sum_{i=1}^{N-1} pb_{ij}^k + \alpha_k z_j$	for all k and J	(33.14)
$t_{ij} = \sum_{i'=1}^{i-1} rt_{ij}^{i'} + \sum_{k=1}^{K} pt_{ij}^k$	for alii and j	(33.15)
$b_{ij} = \sum_{i'=i+1}^{N-I} rb_{ij}^{i'} + \sum_{k=1}^{K} pb_{ij}^k$	for all i and j	(33.16)
$\sum_{ord(j)=1}^{i} xt_{ij} = 1$	for all i	(33.17)
$\sum_{ord(j)=i+1}^{N} xb_{ij} = 1$	for all i	(33.18)
$T_i = \sum_{ord(j)=1}^{i} t_{ij}$	for all i	(33.19)
$B_i = \sum_{ord(j)=i+1}^{N} b_{ij}$	for alii	(33.20)
$PT_i^k = \sum_{ord(j)=1}^{i} pt_{ij}^k$	for all i and k	(33.21)
$PB_i^k = \sum_{ord(j)=i+1}^{N} pb_{ij}^k$	for all 1 and k	(33.22)
" " •-S/	for all i and i' < i	(33.23)
$RB_i^{i'} = \sum_{ord(j)=i+1}^{N} rb_{ij}^{i'}$	for all i and i' > i	(33.24)

Equations (15-18) forty. rt_{rj}^{1} and pt_{y}^{k} in terms of xtg and the total flow of its respective stream.

Equations (15-18) for by. rbq^1 and pb_{tl}^k in terms of xb_{ij} and the total flow of its respective stream.

S,.T,, B,. F,. RT,'. RB,^{f}. PT,^{k}. PB,^{k}, Ok. f$_{tt}$. xs,j. xU_t. xb$_{(J}$ SO

Sg. t,j. b,j pti,^{k}. pb,,^{11}. rt,,$^{1'}$. rb,,$^{1'}$ ≥ 0

It is not necessary to Include equations (15)-(18) for the inlet component flows to the separator, Sq, since the variables xs,j only appear in these equations. Also, the component flows, s,j, only appear in mixers and sharp separators units which can be exactly represented in terms of the individual component flow equations (33.5). (33.7) and (33.9). Equations (33.15)-(33.16) that are the component mass balances for the splitters of top and bottom streams have been included accordingly to the reformulation previously presented. Equations (33.19M33.24) relate the total flow and the individual component flows for the splitter streams.

Preprocessing

Hie proposed superstructure (Fig. 9) allows to bypass certain amount of the feed to the product k, a*, without having to go through the separation network. The amount of the product k that is not bypassed has to be processed in the separation network and it will be denoted as the 'residual product*. Hence, the total 'residual product k' is given by (P^k - a_k) and has the component flows given by (p^ - o^z,)(see Fig. 10).

The global optimal solution of model (32) is a network in which all the 'residual products'.have at least one component with a zero flow. The reason that it is not optimal to separate a stream in the network and later to remix it. The same degree of separation can be achieved using a bypass that does not incur any cost in the objective function.

Consider the second separator in the solution obtained by Floudas (1987) to his second example (see Fig. 11). For this subnetwork of the complete structure the upper 'residual product* has components B and C present. The components are being separated and remixed again. The same outlet flows can be obtained with a smaller input flow to the separator as it is shown in Fig. 11. Note that both 'residual products' have components with zero flow.

It should be clear that if there was not a component with zero flow in the 'residual product[1], then there is part of this stream that could have been obtained by just bypassing the network. This in turn does not incur in any cost, whereas going through the network has a positive cost. The above condition gives a lower bound for the bypass to each product. This also corresponds to the largest amount that can be bypassed since all the flows in the 'residual flow* have to be positive. In this form the bypass can be precalculated without affecting the global optimality of the solution.

The bypass to product k is given by the maximum amount that can be sent to product k without having a negative flow; that is.

a„ = min, [^4 (34)

where Zjis the feed composition and p_{kj} is the flow of component j in product k. The component flows for 'residual product' k. Tkj are given by.

$$\gamma_{kj} = p_{kj} - \alpha_k z_j \qquad (35)$$

Key component bounds

Wehe and Westerberg (1987) proposed using lower bounds for the (low of the key components in separator L These bounds are based on the fact that separator i is the only unit that can perform the task of separating component number 1 from component nutaber 1+1. They are redundant for the nonlinear formulation (32) but they are relevant for the linear relaxation in (33). To calculate them, it is necessary to determine in each product what is the difference between the two key components of separator i with respect to the concentrations in the feed. The lower bounds in separator i' for the flow of the key components in the top (component Jl) and bottom (component J2) streams are given by:

$$t_{ij1} \geq \sum_{k=1}^{K} \{ \gamma_{kj1} - z_{j1} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{for alii} \qquad (36)$$

$$b_{ij2} \geq \sum_{k=1}^{K} \{ \gamma_{kj2} - z_{j2} \min_{j=j1,j2} [\frac{\gamma_{kj}}{z_j}] \} \qquad \text{foralli} \qquad (37)$$

where Ykj is given by (35). It is important to include both bounds in the relax model (33) since there is no guarantee that the inlet flow to separator i has the same proportion between the key components as the feed. It is not known in which part of the sequence separator i will be placed, and it can be after a splitter that is not being approximated correctly.

The bounds in (36) and (37) can be extended to separation of components that are not adjacent in the feed. Consider component number i and component number i+3. There are three separators that can perform this task/separators i. i+1 and i+2. Cuts of the following form can be obtained.

t_t i + t_M i + t|*a 12 £ {.Dd - zt min_ord(j),< or H3 [-r]} (K above £; γki above r) for all i (38)

b_i1 + b_i+1 i + b_M i 2 £ {Tki*3 - z,*_3 minors_OM *3 i~)) (K above £; ykl above i~) for all i (39)

Equations as the ones in (38) and (39) can be redundant compared to equations (36) and (37) and it is possible to detect this before solving the problem.

Relativeflowrate constraints

These constraints are used when the relation between the flowrates of two components is known. In particular, consider component A in the last column of the network (see Fig. 12). None of the redirected streams contains component A. Therefore, the relative flowrate of component A with respect to the other components in the top stream has to be smaller than in the feed. This relation should remain valid after the top stream is split to the products and redirected flows.

In the separator previous to the last one, N-2, all the streams do not have component A except the one coming from the top of the last column. For this one it is already known that the relative flow of component A with respect to the other components is smaller than in the feed. This type of analysis can be done for component A and component N in all the columns yielding the following linear constraints for the splitters.

for all i and k

$$z_A\, pt_{ij}^{k} - z_j\, pt_{iA}^{k} \geq 0 \qquad \text{for } j \neq A \text{ and ord}(j) \leq i \qquad (40)$$

$$z_N\, pb_{ij}^{k} - z_j\, pb_{iN}^{k} \geq 0 \qquad \text{for ord}(j) \neq N \text{ and ord}(j) > i \qquad (41)$$

for all i and i' > i

$$z_A\, rt_{ij}^{i'} - z_j\, rt_{iA}^{i'} \geq 0 \qquad \text{for } j \neq A \text{ and ord}(j) \leq i \qquad (42)$$

for all i and i' < i

$$z_N\, rb_{ij}^{i'} - z_j\, rb_{iN}^{i'} \geq 0 \qquad \text{for ord}(j) \neq N \text{ and ord}(j) > i \qquad (43)$$

Bounds on concentrations and total flows

The approximations (15-18) require bounds for the total flows and component concentrations in the splitters. The lower bound for the total flow of the top and bottom streams is given by the lower bound of the key components obtained in equations (36) and (37). For the outlet streams of the splittters, that are the redirected streams and the streams that go to the products, the lower bound is zero. The upper bound for the total flow of the top and bottom streams is given by the feed to the network of the components that can be present in each stream .that is.

$$T_i^U = \sum_{ord(j)=1}^{i} Feed \cdot z_j \qquad \text{for all } i \qquad (44)$$

$$B_i^U = \sum_{ord(j)=i+1}^{N} Feed \cdot z_j \qquad \text{for all } i \qquad (45)$$

The upper bound for the streams after the splitter are given by.

$RT_i^{i'u} = T_i^u$ for all i and i' < i (46)

$RB_i^{i'u} = B_i^u$ for all i and i' > i (47)

$PT_i^k = \sum_{and(j)=1}^{i} Y_{kj}$ for all i and k (48)

PB,[k] = Jb? for all i and k (49)

The lower bounds for the concentrations are zero except for fee key components in the separator for which the lower bounds are given by the lower bound of its flow divided by the upper bound of the total flow of that stream. The upper bounds in the concentrations are given by one minus the lower bounds of the other components.

The solution of the linear programming model (33) provides a lower bound to the global optimum since this model is a valid relaxation of the nonconvex model (32). This lower bound is obtained by solving the LP model for the 'residual products' in (35) with the addition of the valid constraints (36)-(43).

The upper bounds are generated solving model (32) in terms of the individual flows for the 'residual products'. When additional nonconvex problems are solved to improve the current upper bound it can happen that very similar initial point are generated. In this case, a new nonconvex NLP is solved in which bounds over the total inlet flows to the separators (S_i) are included. For this purpose the values of these variables in the LP solution (S_i^*) arc used such that the current incumbent solution is no longer feasible.

Example4

Consider the 3 component example proposed by Floudas and Aggarwal (1990). An equimolar feed has to be separated into two products as show in Fig. 13. The objective function is given by

$$\Phi = 0.2395 + 0.00432\, S_1 + 0.7584 + 0.01517\, S_2 \qquad (50)$$

The bypass to products 1 and 2 can be calculated according to equation (34) and the 'residual product' component flows are obtained through equation (35) (see Fig. 14). The total bypass to product 1 is $\alpha_1 = 90$ and the bypass to product 2 is $\alpha_2 = 150$ and the feed has a concentration of $z_A = 1/3$, $z_B = 1/3$ and $z_C = 1/3$. In this form the 'residual product' 1 is $Y_{1A} = 0$, $Y_{1B} = 20$ and $Y_{1C} = 0$ and the 'residual product' 2 is $Y_{2A} = 20$, $Y_{2B} = 0$ and $Y_{2C} = 20$. Additionally, lower bounds on the flow of the key components in both separators are obtained

using equations (35M36). The key components in separator 1 are component A in the top and its flow has to be at least 20 and component B in the bottom has to have at least a flow of 20. In the top stream of the second separator at least 20 units of component B have to be separated from 20 units of component C in the bottom stream. It is important to note that after preprocessing the network several suboptimal solutions have been cut off. One of these suboptimal solutions for this particular data is a parallel configuration of both separators (there are situations in which a parallel configuration can correspond to the global solution as will be shown in example 5). In this example the direct or indirect sequence have a lower objective function. Both of these configurations are local solutions with an objective function value of 4 = 1.8639 for the direct sequence and $ = 2.081 for the indirect one. In some instances, MINOS 5.2 had problems converging even in this small example.

The LP (33) is formulated for this problem, giving a lower bound of ^ = 1.8639. The approximations are exact and therefore this solution is a feasible solution of model (32) proving that it corresponds to the global optimum. Hence, convergence is achieved in one iteration. The optimum solution corresponds to the direct sequence shown in Fig. 15. It should be noted that if the linear mass balances for the mixer for product 2 were not considered since they are redundant for the nonlinear formulation, a lower bound in the relaxed model of $\$_L$ = 1.12 is obtained. This shows that It is relevant to include all the mass balances in the linear model in order to tighten the lower bound.

ExompteS

In the approach proposed by Wehe and Westerberg (1987) for the case of 3 components only the direct and indirect sequences are considered and both options can be modelled as LP problems since no mixing is required for these separation networks. However, this example shows that parallel configurations can be also globally optimal and that they are not excluded by the method proposed in this paper. To be able to consider parallel configurations or any combination of parallel with direct or indirect sequences it is necessary to model a superstructure in which mixing is allowed (like in the structure used in Fig. 13). Here, nonconvexities arise in the mass balance equations after the separators.

Consider that an equimolar feed is to be separated into the two different products given in Fig. 16. The objective function is to minimize the sum of the total flows into the separators. The same procedure that in the previous example is followed and the bypass can be precalculated by equation (34). The solution to the model (32) yields Φ_L = 12 and since it is a feasible solution of model (32) it corresponds to the global optimum (see Fig. 16). Note that the solutions for the direct or indirect sequences have an objective function of $ = 16.

Branch and Bound

If there is a gap between the lower ai}d upper bound, a branch and bound search is performed. It is only necessary to do the search over the variables involved in the nonconvex terms. The concentrations are used as the branching variables since a change in them affects the inlet and outlet streams of a splitter. In this way. it is necessary to check the approximation for the concentrations in the splitters of the top and bottom strear^s of the separator. Equations (26)-(29) for the splitters of top and bottom streams are used to perfom the branch and bound search.

Results

Table 1 summarizes the results of the earlier examples 1 to 3 and of the sharp separation network examples 4 to 12. The number of variables is the total number of variables that are included in the reformulated and relaxed model (33) for that example. The lower bound is the initial bound that is obtained by solving model (33) over the entire feasible space. The inital gap represents the percentage difference between the initial lower and upper bounds. When there is a zero initial gap it means that the first relaxed solution is feasible in the original problem thereby corresponding to the global solution. The column for nodes gives the total number of subproblems that where solved before converging to the global solution. A relative tolerance of 0.01 was used, except for example 2 were exact convergence was obtained after branching and example 12 for which a tolerance of 0.02 was used. It is important to note that the initial lower bound is tight and that it corresponds to a good estimation of the global solution. The largest differences are for example 1 with a 25% of difference and for example 12 with a 7% difference. The LP time refers to the time used to solve each relaxed model and the NLP time is the time used for solving a nonconvex model. It is possible to do updates using the previous LP solution and in this form have a more efficient implementation. The times are in seconds and the problems were solved on an IBM RS600/530 using GAMS 2.25 (Brooke et al. (1988)). MINOS 5.2 was used to solve the LP problems and CONOPT for the nonconvex NLP problems. A brief description of the example problems 6 to 12 is given below. It includes the specific data for the problem, the objective function and the topology of the network that is the global solution.

Example 6

This example corresponds to example 2 from Floudas (1987). In this case a linear objective function with the same cost coefficients is used and it is given by,

$$(M2.5S!+3.0S_2+1.5S_3 \qquad (51)$$

The data for the composition of the products is given in Table 2.

The initial lower bound is ϕ_L = 54.25 and an upper bound of $ = 55.5 is obtained by solving the nonconvex problem. A partition of the feasible region is performed using the composition of component D in the bottom stream of separator 1. The first subproblem (x £ 0.166) yields a lower bound of ^ = 55.45 and the second subproblem (x < 0.166) has a solution of ϕ_L= 55.8. The latter is greater than the upper bound and the former is less than 1% of the global solution (see Fig. 17).

Exampie7

This example is taken from Floudas (1987). The data for this problem is given in Table 3 and the linear objective function is given by:

$$\phi = 2.5\,S_1 + 3.0 S_2 + 1.2 S_3 \tag{52}$$

The initial lower bound is ϕ_L = 32.7 and it provides a feasible solution to the nonconvex problem. In this form the global solution (see Fig. 18Ms obtained in one iteration. It is interesting to see that this solution also provides a better objective function for the concave objective function used by Floudas (1987) (4 = 10.65 versus 0 = 13.68 which is 28% higher)

Elxample8

This four component problem is taken from Wehe and Westerberg (1987). The data for the products is given in Table 4 and the objective function has the following form:

$$\phi = 5.0 + 0.5\,S_1 + 4.0 + 0.3\,S_2 + 6.0 + 0.7\,S_3 \tag{53}$$

The first relaxed subproblem has a solution of ϕ_L =26,76 and it is infeasible for the nonconvex problem. A nonconvex problem is solved using CONOPT with the LP solution as the initial point. An upper bound of 0 « 26.79 is obtained corresponding to the global solution (see Fig. 19) within a 0.1%.

Example 9

This example corresponds to example 1 from Wehe and Westerberg (1987). Table 5 provides the data for the product flows and the objective function is given by:

$$\phi = 5.0 + 0.5\,S_1 + 9.0 + 1.0\,S_2 + 3.0 + 0.4\,S_3 + 6.0 + 0.6\,S_4 \tag{54}$$

A initial lower bound of ϕ_L = 85.16 is obtained and the upper bound Is 0 » 85-65. The difference is 0.5% and the global solution (see Fig. 20) is obtained in one iteration.

Example 10

This problem is taken from Flotidas (1987) and the data is given in Table 6. The objective function is given by,

$$\Phi = 1.2\,S_1 + 3.0\,S_2 + 2.5\,S_3 + 1.5\,S_4 \qquad (55)$$

The initial lower bound is Φ_L = 156.56 and the upper bound is Φ^U = 179.08. After 5 nodes the global solution of Φ = 159.48 (see Fig. 21) is obtained.

Example 11

The data for this 6 component problem are given in Table 7 and the objective function has the following form:

$$\Phi = 1.5\,S_1 + 3.0\,S_2 + 2.0\,S_3 + 1.0\,S_4 + 4.0\,S_5 \qquad (56)$$

The initial lower bound is Φ_L = 173 and the upper bound is Φ^U = 179.11. After five nodes the global solution is obtained (see Fig. 22).

Example 12

This is a 6 component 4 products problem and the data are given in Table 8. The objective function is:

$$\Phi = 5.0\,S_1 + 3.0\,S_2 + 2.0\,S_3 + 2.5\,S_4 + 4.0\,S_5 \qquad (57)$$

The initial lower bound is Φ_L = 362 and the initial upper bound is Φ^U = 415.6. The global solution of Φ = 388 (with a 2% optimality gap) is obtained after 33 nodes (see Fig. 23).

Conclusions

A general procedure for the global optimization of process networks with multicomponent streams has been proposed. The basic idea relies on a relaxed LP model that is obtained through reformulation-linearization techniques that establish a clear relation between the component flow and the composition models for mass balances. The reformulated model combines both of these providing tighter lower bounds than other relaxations proposed in the previous work. The relaxed linear model has been embedded in a branch and bound procedure to obtain the global optimal solution.

As has also been shown, the algorithm can be further specialized to take advantage of the particular structure of sharp separation networks with single feed and mixed products. Here, it is possible to preprocess the problem to reduce the space over which the search is

conducted. The bounds that are necessary for the estimator functions in the relaxed model can be obtained without having to solve any subproblems. Different types of linear approximations that are nonredundant to the relaxed model are included to obtain a tighter lower bound.

Twelve examples for both general process networks and for sharp separation networks have been presented to illustrate the performance of the algorithm. As has been shown, only a small number of nodes are commonly needed in the branch and bound search to identify the global or e-global solution. Moreover, in many cases the initial lower bound is either the exact solution or a very good approximation to the global solution.

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Appendix A. Reformulation-Linearization to obtain the nonlinear constraints in **model MF**

The nonlinear equations in **model** MF, that can be expressed either as (8) or (9), can also be generated from model MX. For this purpose take the concentration model MX and consider equation (5),

$$x_j^k = x_j^l \qquad \text{(A.1)}$$

multiply by the valid bound constraint Xj-k £ 0

xfxfmxfzp **(A.2)**

Use equation (5) for **component** j\

x,kVil*fk** **1A-3'**

Multiply by **the valid bound** constraints $F^k \geq 0$ and $F^1 \geq 0$,

F^JP^-F^F*^ tA.41

that it is linearized to yield.

fffJJsfj'ff (A.5)

which is precisely equation (8) for the splitters in the individual flow model MF.

Consider again equation (5).

$$x_j^k = x_j^1 \qquad \text{(A.6)}$$

multiply by the valid bound constraints $F^k \geq 0$ and $F^* \geq 0$,

$$F^k x_j^k F^l = F^l x_j^l F^k \qquad \text{(A.7)}$$

that can be linearized to yield,

ffF^f/F* (A.8)

Define the split fraction ? to be,

?=|* (A.9)

Equation (A.8) can then be expressed as

f/ = ? fjk (A.10)

which corresponds to equation (9).

Hence, the nonlinear equations (8) and (9) are redundant to model MX. Their linear approximations in general are also redundant in the linear reformulated model MR Consider equation (A.1O), similarly to (15) one of its linear approximations is given by.

$$f_j^l \geq \xi^{lL} f_j^k + \xi^l f_j^{kL} - \xi^{lL} f_j^{kL} \qquad \text{(A.11)}$$

If there are no particular restrictions in the splitters, then the bounds for the split fraction variable are $0 \leq \xi \leq 1$ and using them in (A. 11) yields.

$$f_j^1 \geq f_j^{kL} \xi^1 \qquad \text{(A.12)}$$

The bound for the individual component flow is given by $f_j^{kL} = x_j^L F^{kL}$: also $x_j^k = x_j^1$ and $\xi^1 = \frac{F^1}{F^k}$, which leads to.

$$f_j^1 \geq x_j^L F^1 \frac{F^{kL}}{F^k} \qquad \text{(A.13)}$$

The estimator (15) for the same conditions ($F^{*L} = 0$) is given by

$$f_j^1 \geq x_j^L F^1 \qquad \text{(A.14)}$$

Since the factor $\frac{F^{kL}}{F^k}$ is always less or equal than 1, equation (A. 13) is redundant. A similar analisis can be performed for the other estimators. Only when more specific bounds over the split fractions or the individual component flows are known, will these additional estimators be non redundant.

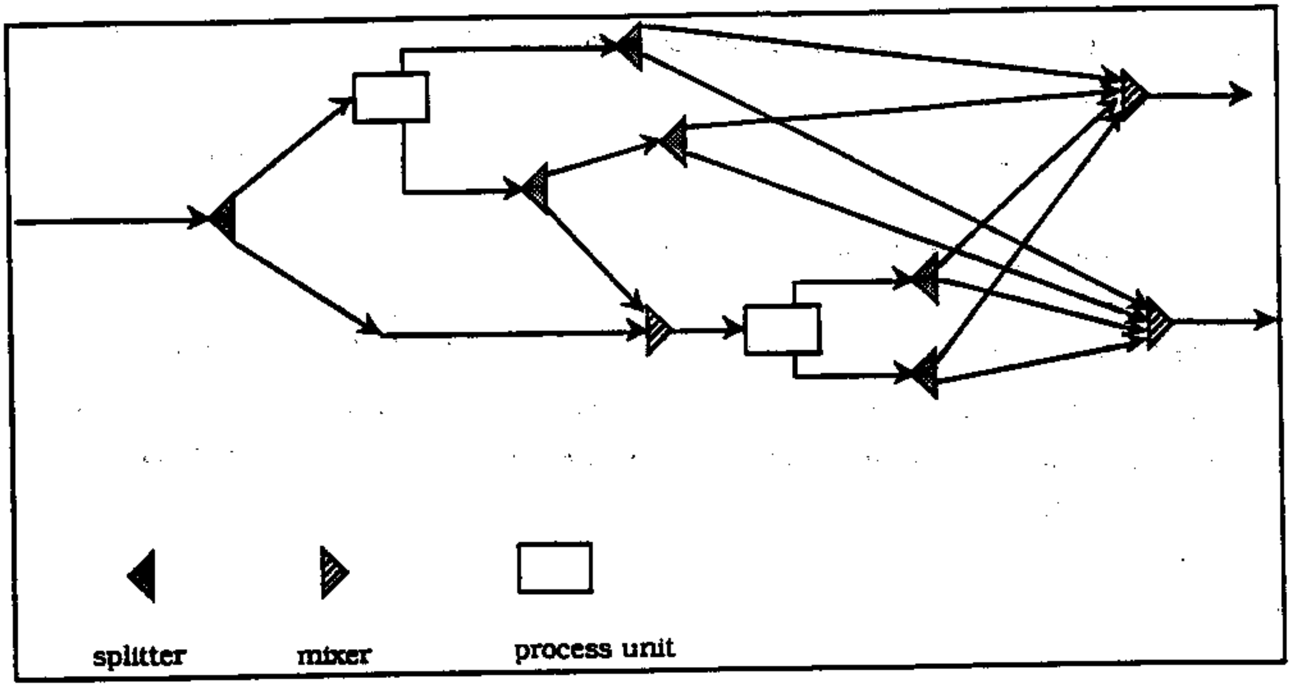


Figure 1. Process network with units, splitters and mixers.

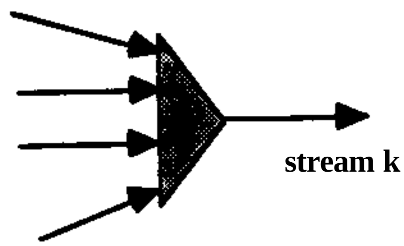


Figure 2. Mixer module.

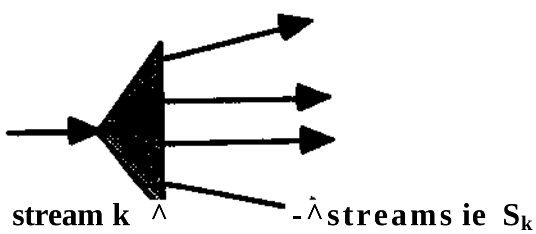
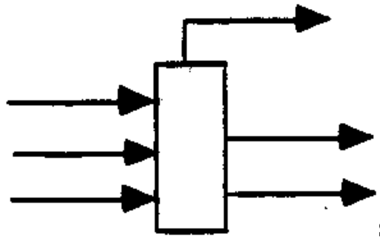
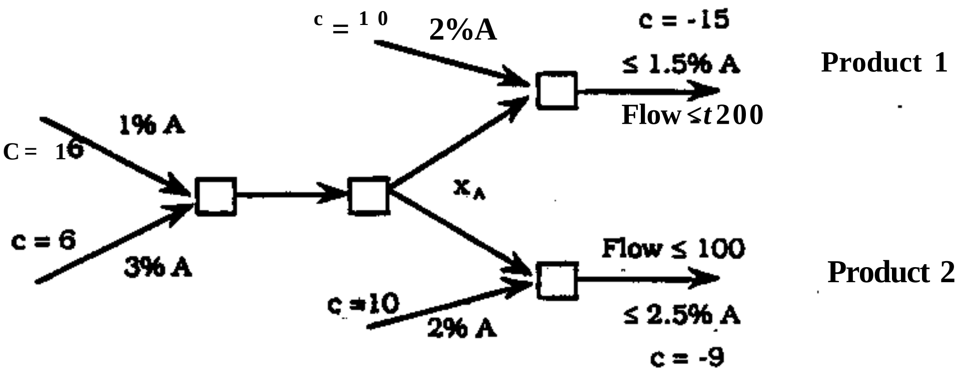


Figure 3. Splitter module.

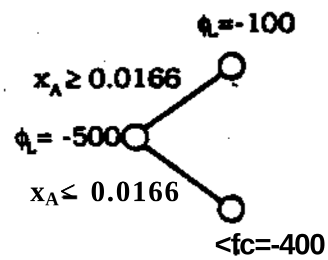


streams $i \in I_k$ streams $i \in O_k$

Figure 4. Process unit module.



(a) Network



(b) Tree search

Figure 5. Network and branch and bound search for example 1

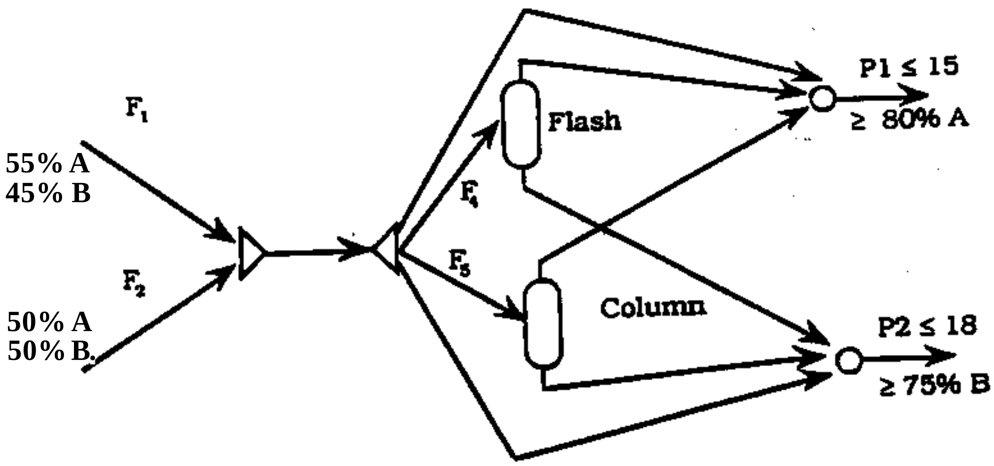


Figure 6. Network for example 2.

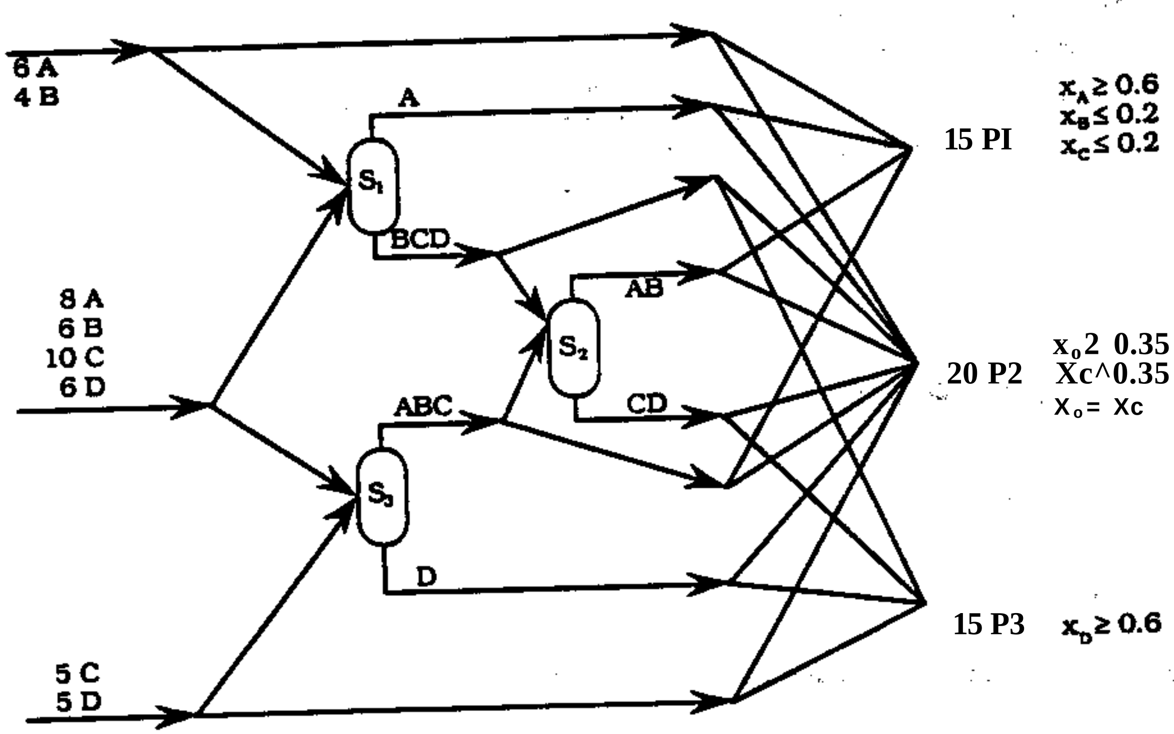


Figure 7. Network for example 3.

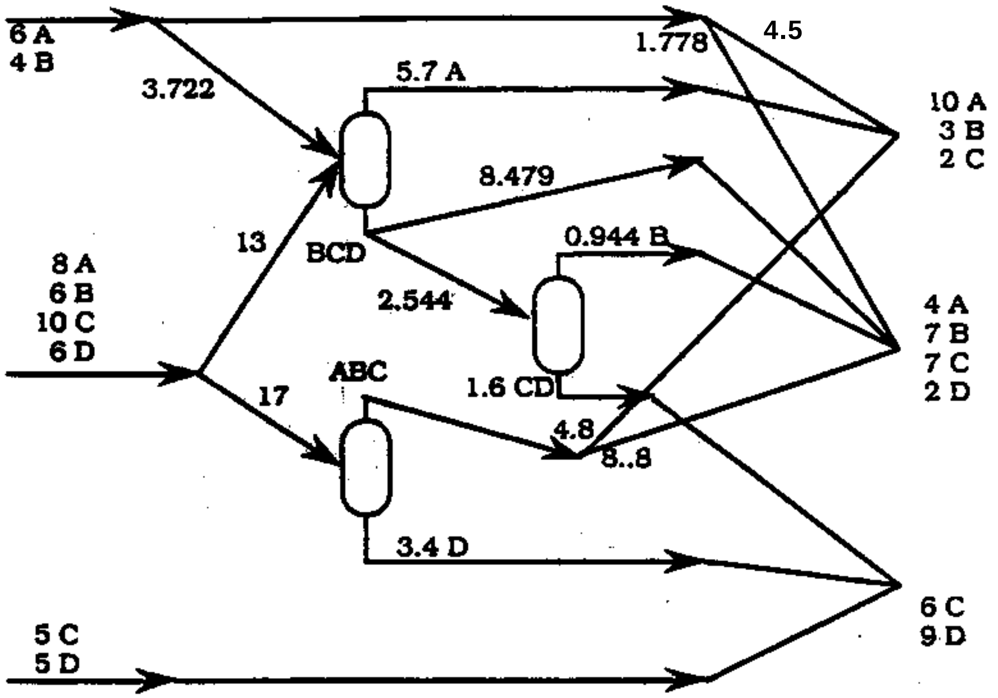


Figure 8. Optimal network for example 3.

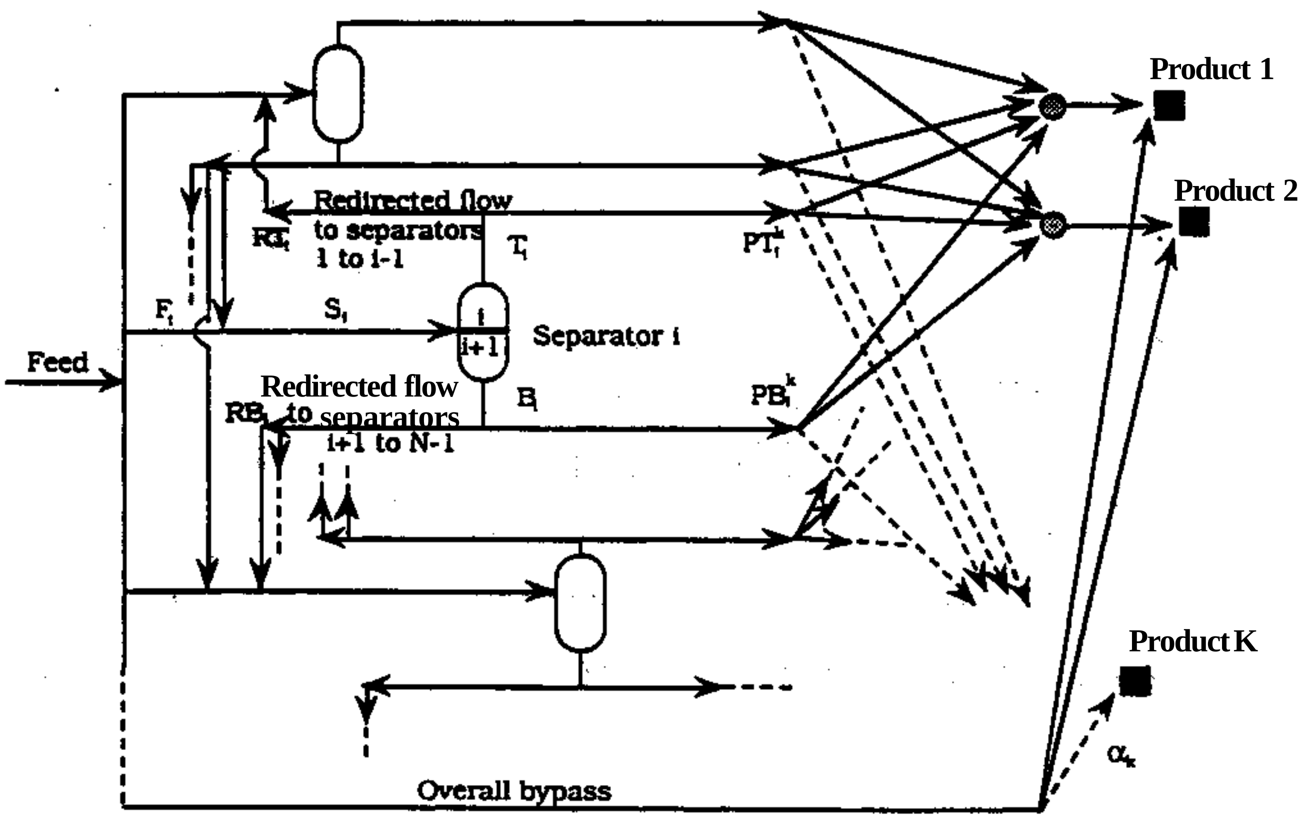


Figure 9. Superstructure for separation with sharp splits and mixed products.

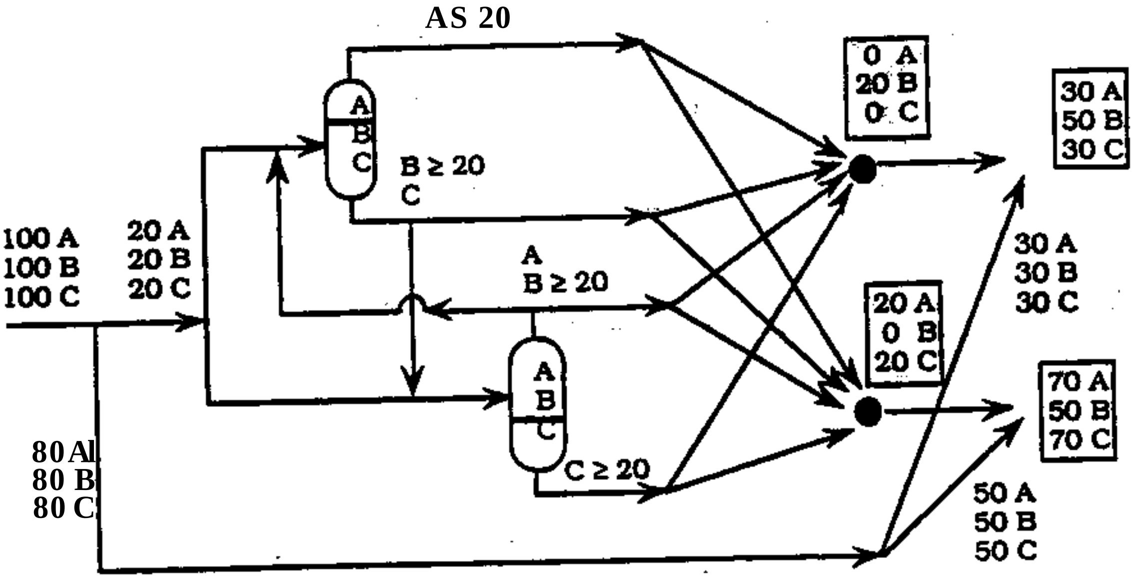


Figure 14. Residual products and key component bounds in example 4.

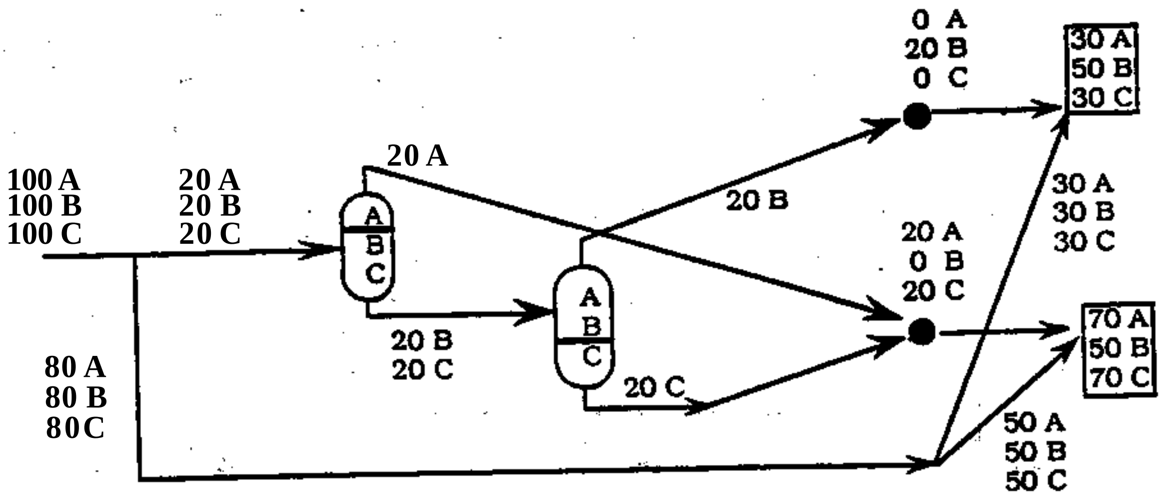


Figure 15. Global optimum solution of example 4.

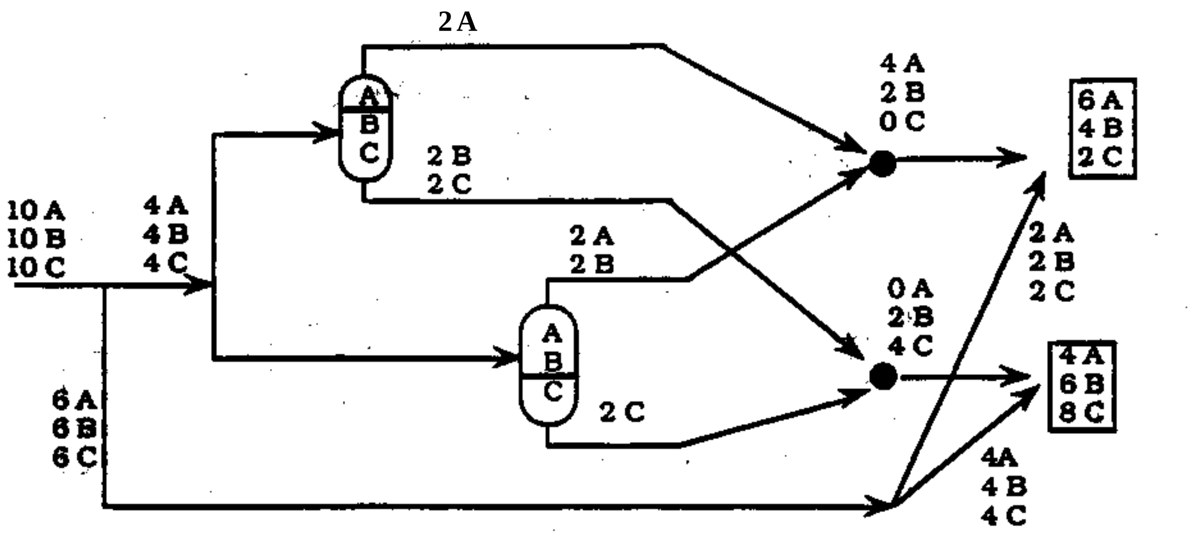


Figure 16. Global optimum solution of example 5.

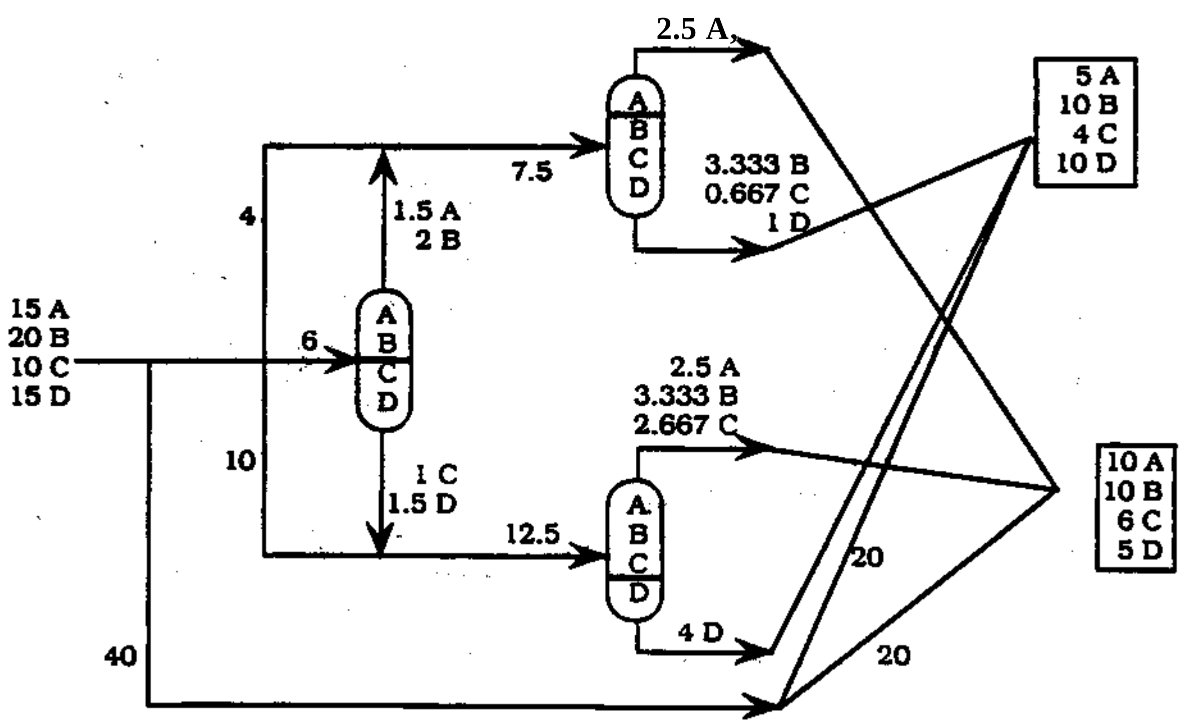


Figure 17. Solution of example 6.

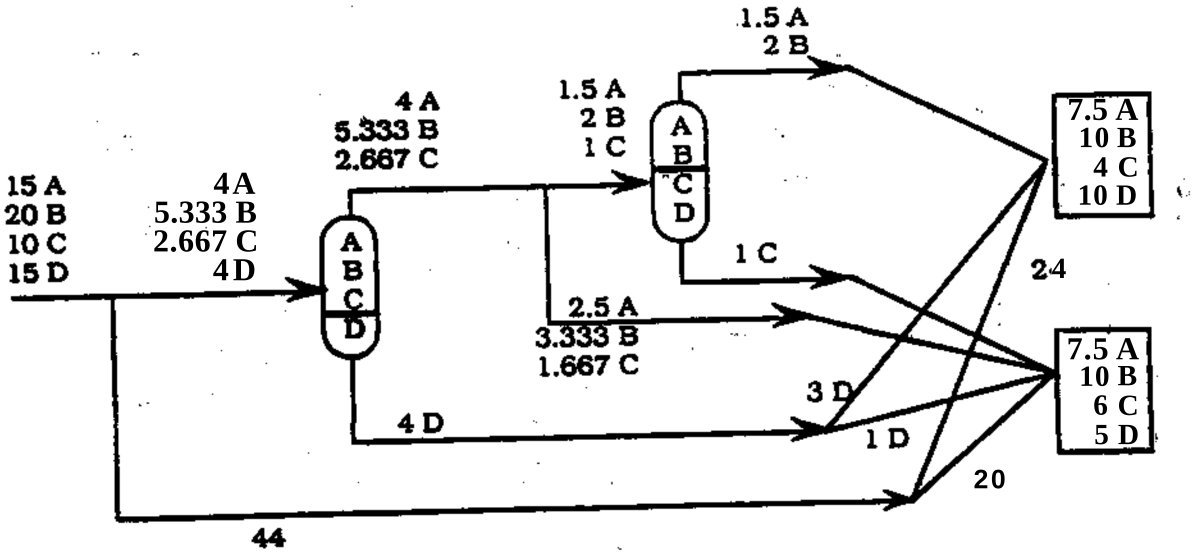


Figure 18. Solution of example 7.

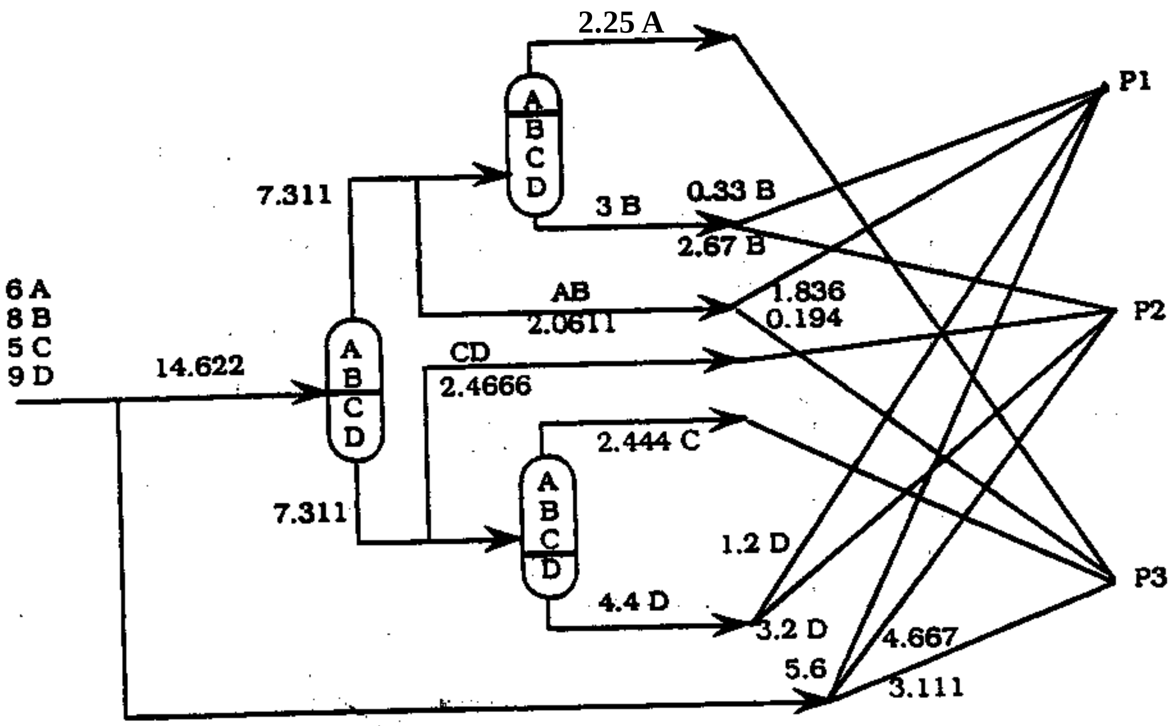


Figure 19. SoluUon of example 8.

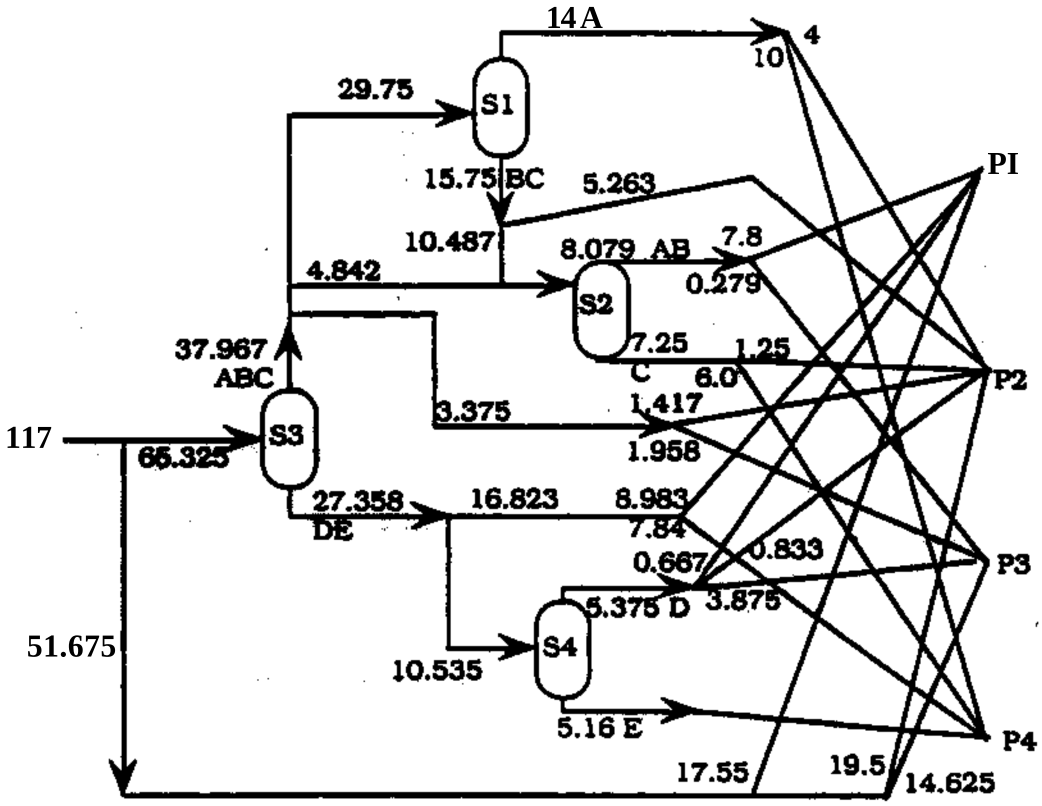


Figure 20. Solution of example 9.

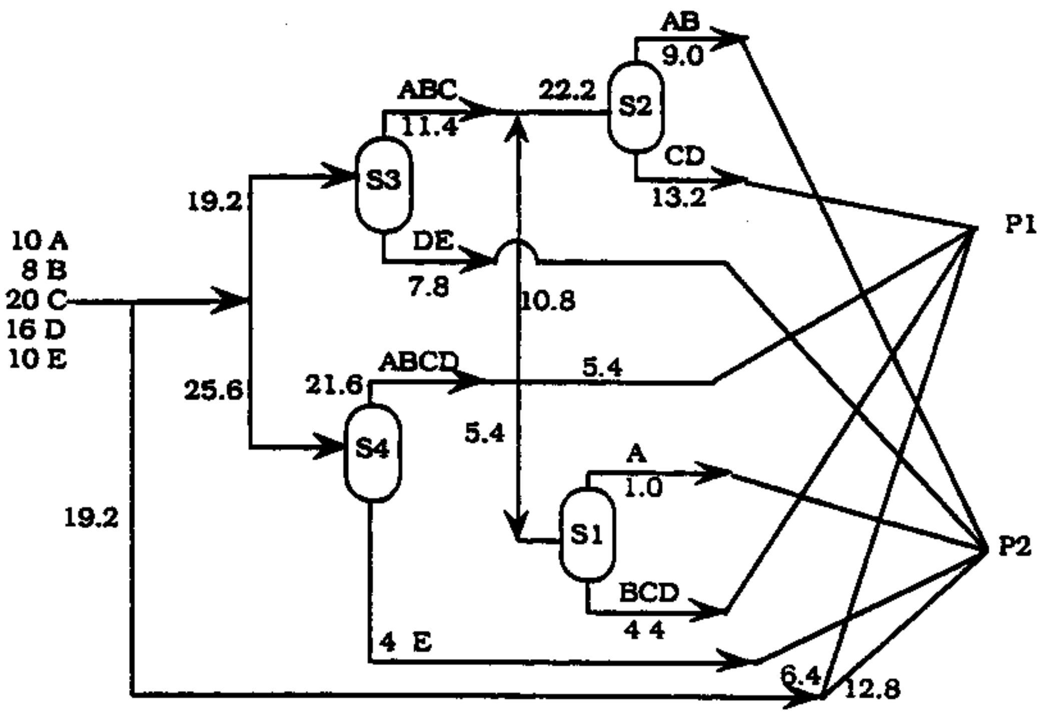


Figure 21. Solution of example 10.

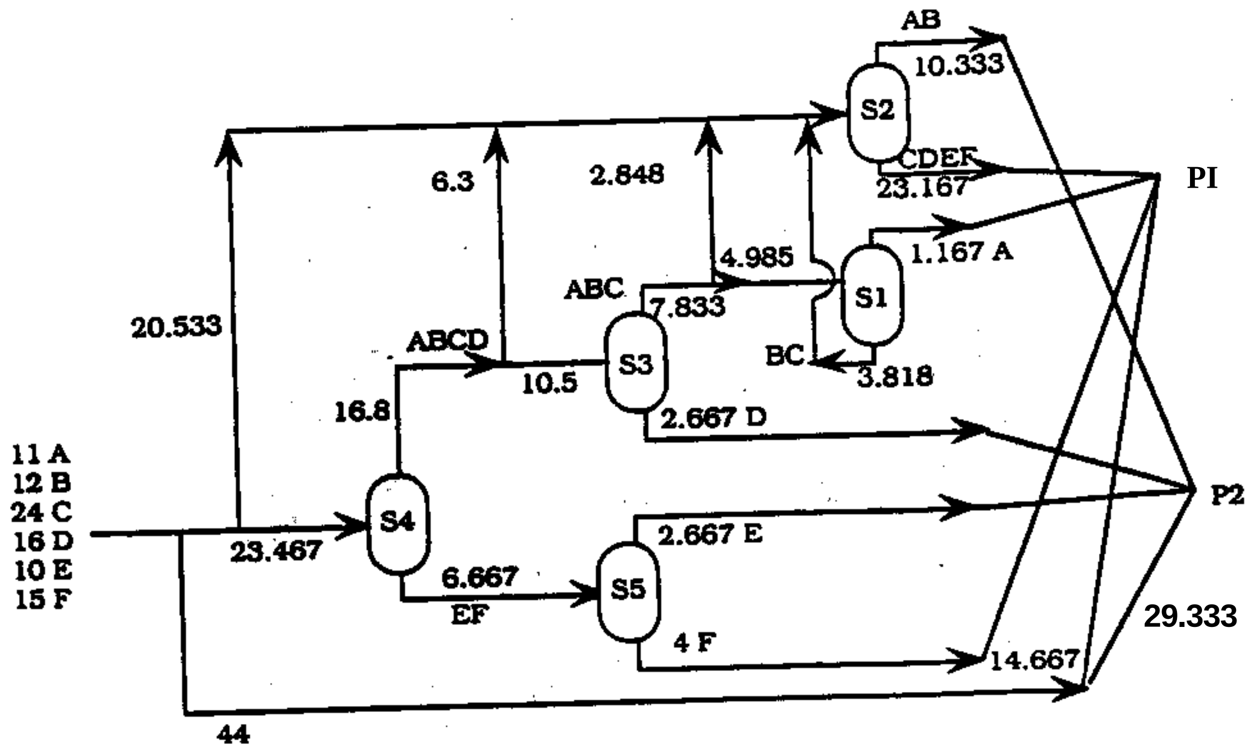


Figure 22. Solution of example 11.

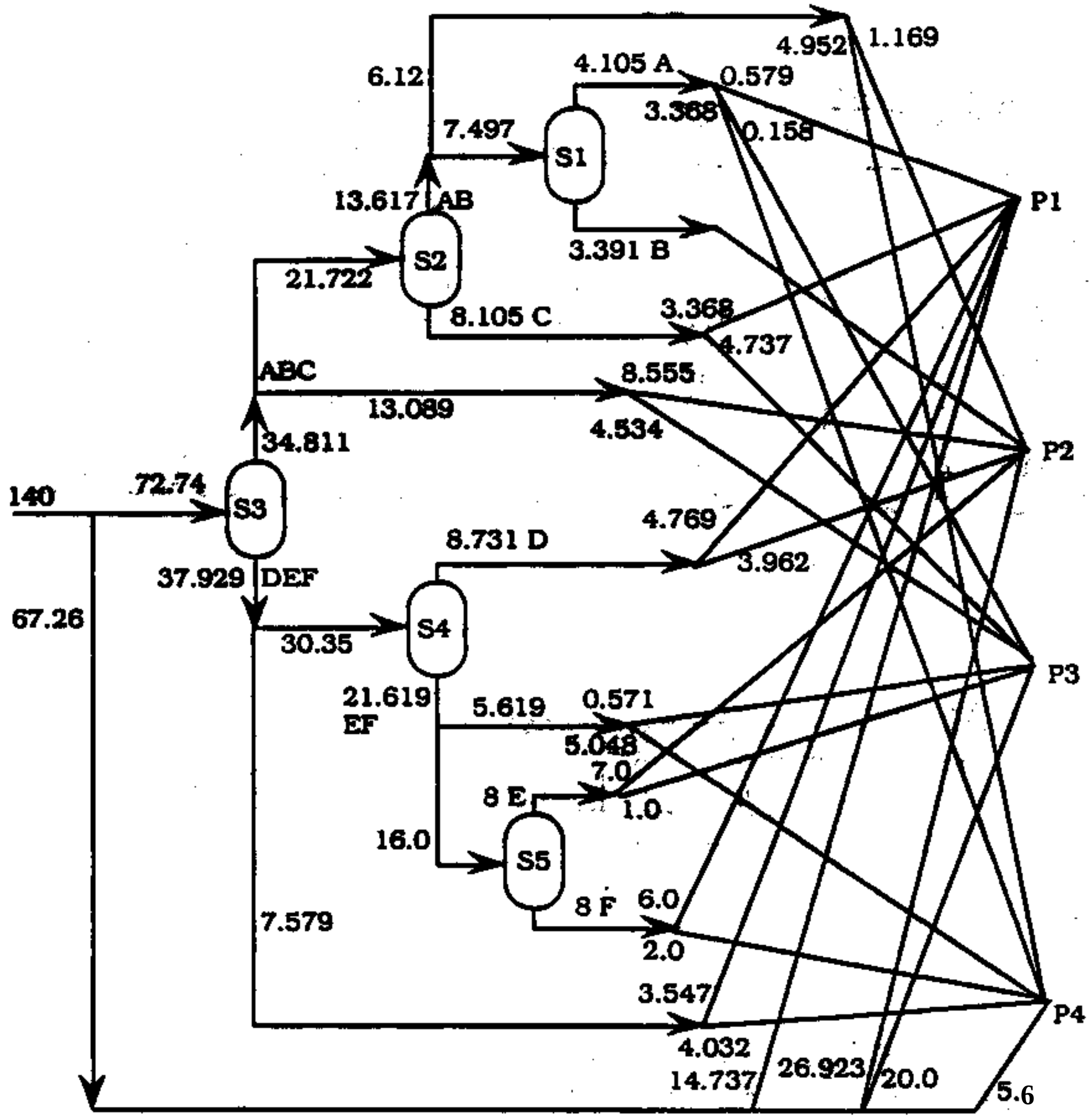


Figure 23. Solution of example 12.

Table 1. Computational results

	Comp.	>rocL	Var.	Lower bound	Initial gap	Global solution	Nodes	LP time	NLP time
Example 1	—	...	29	-500	20	-400	3	0.05	0.1
Example 2	...	...	35	-513.22	0.3	-511.87	3	0.26	0.3
Example 3			113	138.18	0.4	138.7	1	0.34	0.4
Example 4	3	2	65	1.8639	0.0	1.8639	1	0.13	—
Example 5	3	2	65	16	0.0	16	1	0.13	—
Example 6	4	2	107	54.25	2.3	55.5	3	0.97	0.4
Example 7	4	2	107	32.7	0.0	32.7	1	0.17	—
Example 8	4	3	125	26.76	0.1	26.79	1	0.23	0.3
Example 9	5	4	281	85.16	0.5	85.65	1	3.08	2.8
Example 10	5	2	225	156.56	12.4	159.48	5	2.59	2.3
Example 11	6	2	350	173	3.5	179.11	5	9.98	8.8
Example 12	6	4	430	362	14.8	388	33	19.8	13.2

Table 2. Data for example 6.

Component	A	B	C	D	Total
Product 1	5	10	4	10	29
Product 2	10	10	6	5	31
Feed	15	20	10	15	60

Table 3. Data for example 7.

Component	A	B	C	D	Total
Product 1	7.5	10		10	31.5
Product 2	7.5	10	6	5	28.5
Feed	15	20	10	15	60

Table 4. Data for example 8.

Component	A	B	C	D	Total
Product 1	2	3	1	3	9
Product 2	1	4	1	5	11
Product 3	3	1	3	1	8
Feed	6	8	5	9	28

Table 5. Data for example 9.

Component	A	B	C	D	E	Total
Product 1	7	8	3	9	8	35
Product 2	10	3	5	5	4	27
Product 3	5	5	6	7	3	26
Product 4	10	0	6	4	9	29
Feed	32	16	20	25	24	117

Table 6, Data for example 10.

Component	A	B	C	D	E	Total
Product 1	2	2.4	16	8	1	29.4
Product 2	8	5.6	4	8	9	34.6
Feed	10	8	20	16	10	64

Table 7. Data for example 11.

Component	A	B	C	D	E	F	Total
Product 1	3	2	16	8	4	10	43
Product 2	8	10	8	8	6	5	45
Feed	11	12	24	16	10	15	88

Table 8. Data for example 12.

Component	A	B	C	D	E	F	Total
Product 1	3	2	6	8	4	10	33
Product 2	8	10	8	8	6	5	45
Product 3	5	4	10	3	11	4	37
Product 4	7	3	1	2	5	7	25
Feed	23	19	25	21	26	26	140

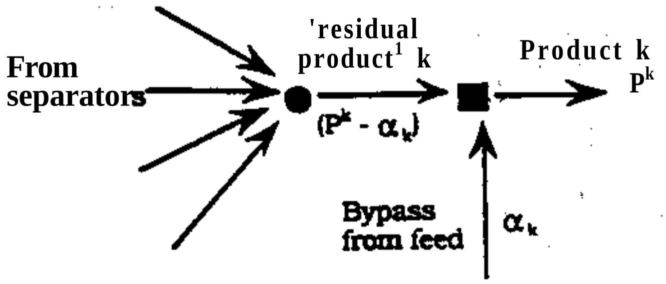


Figure 10. Definition of 'residual product'.

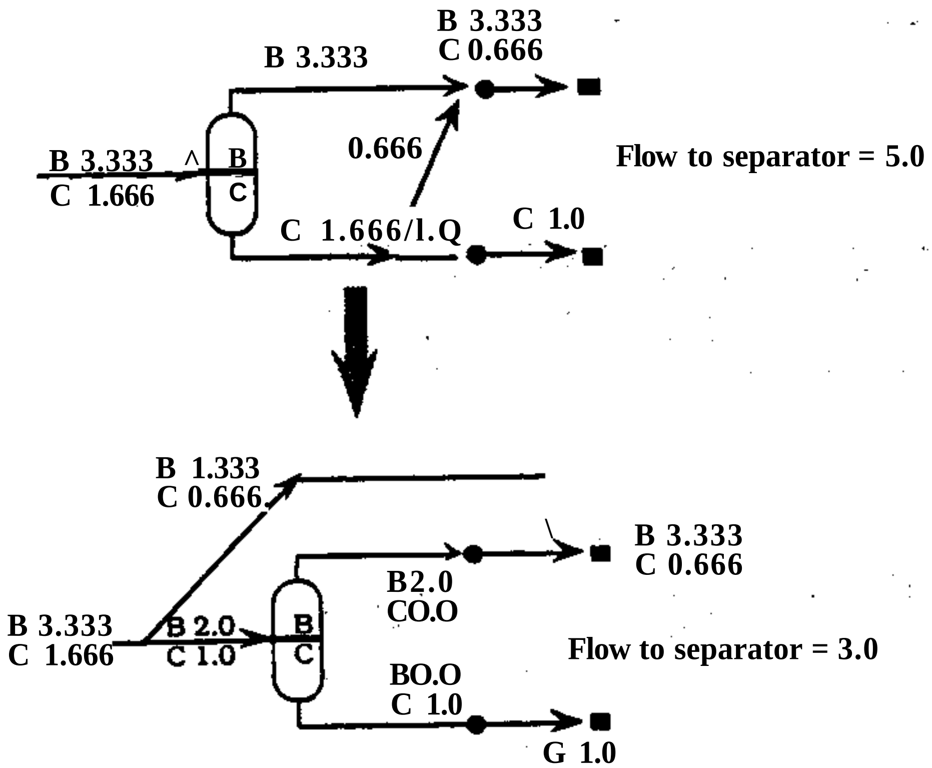


Figure 11. Example of solution without and with a zero component flow in 'residual product'.

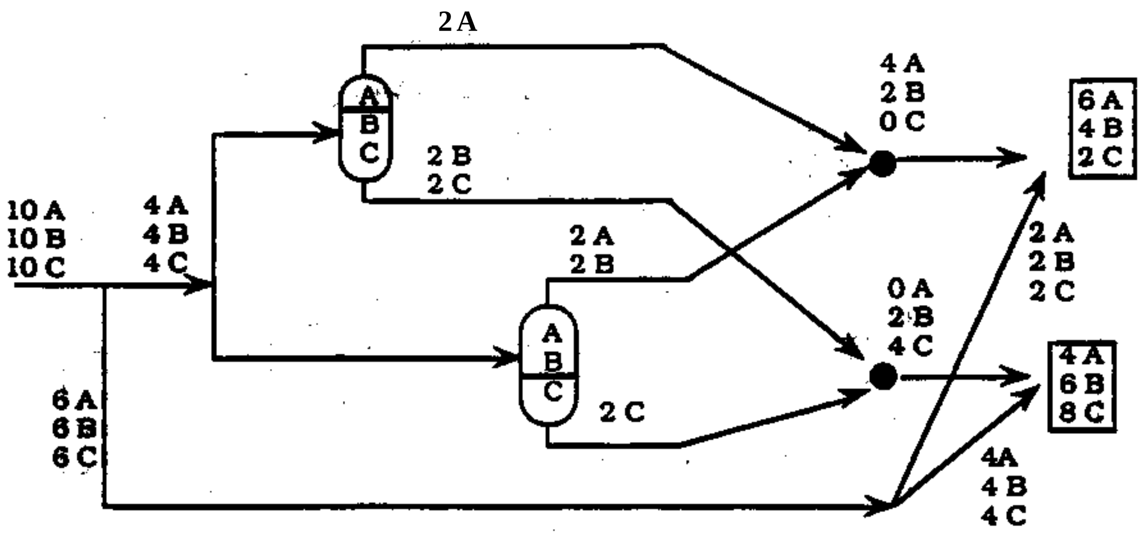


Figure 16. Global optimum solution of example 5.

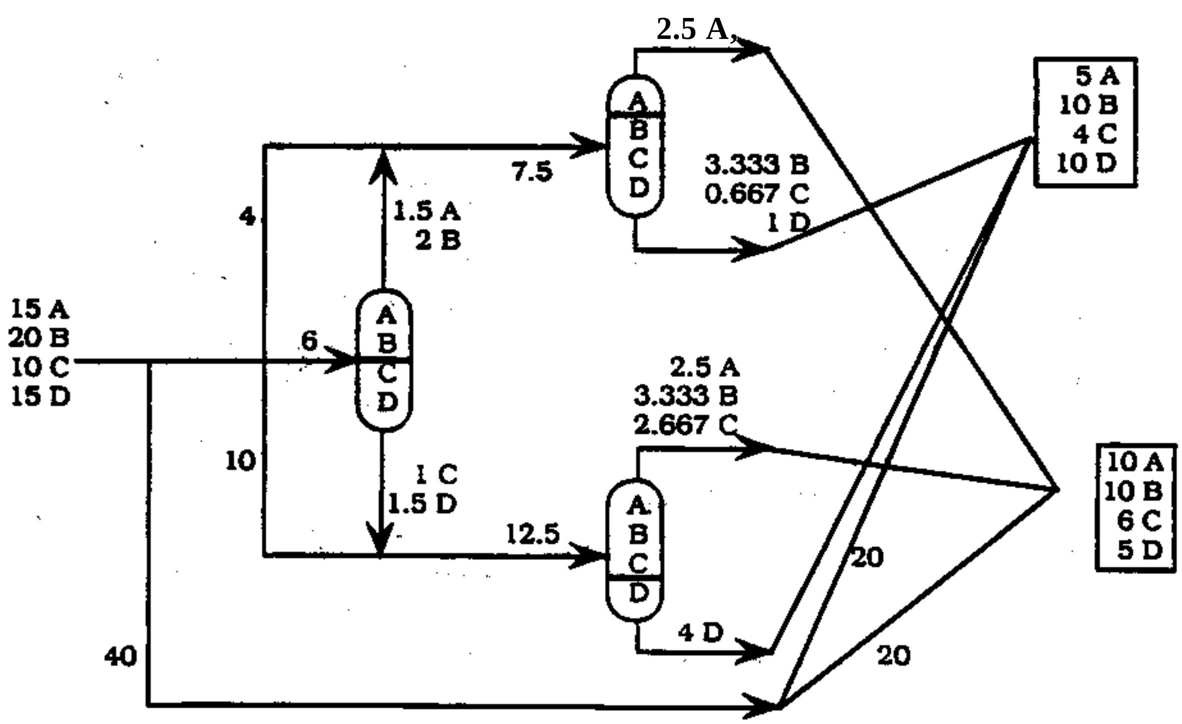


Figure 17. Solution of example 6.

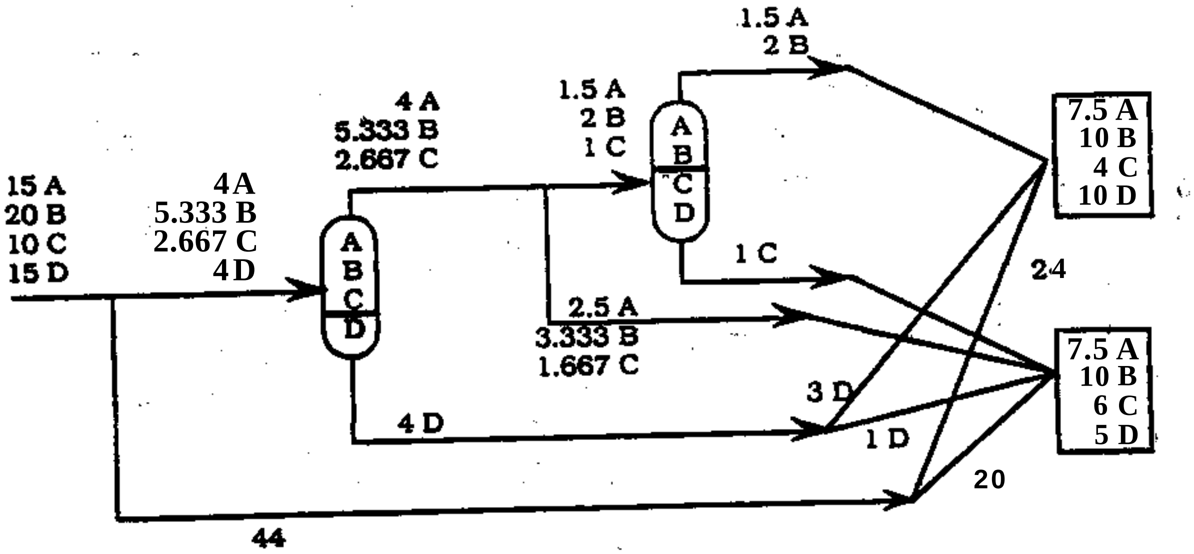


Figure 18. Solution of example 7.

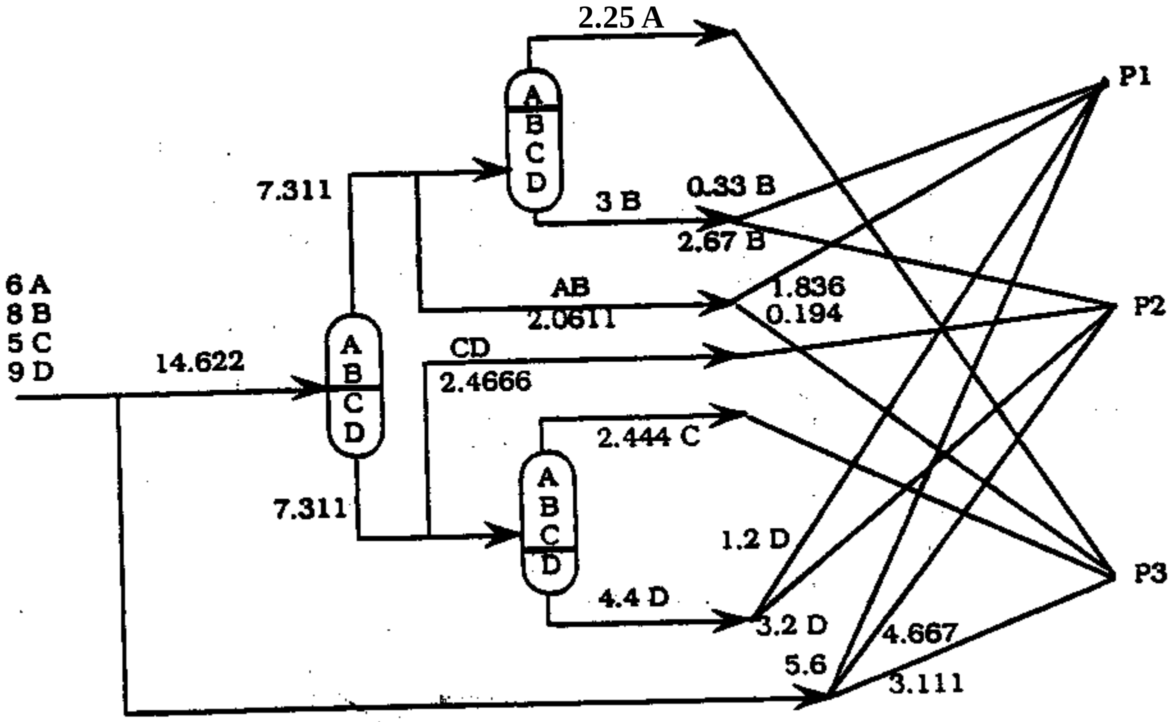


Figure 19. SoluUon of example 8.

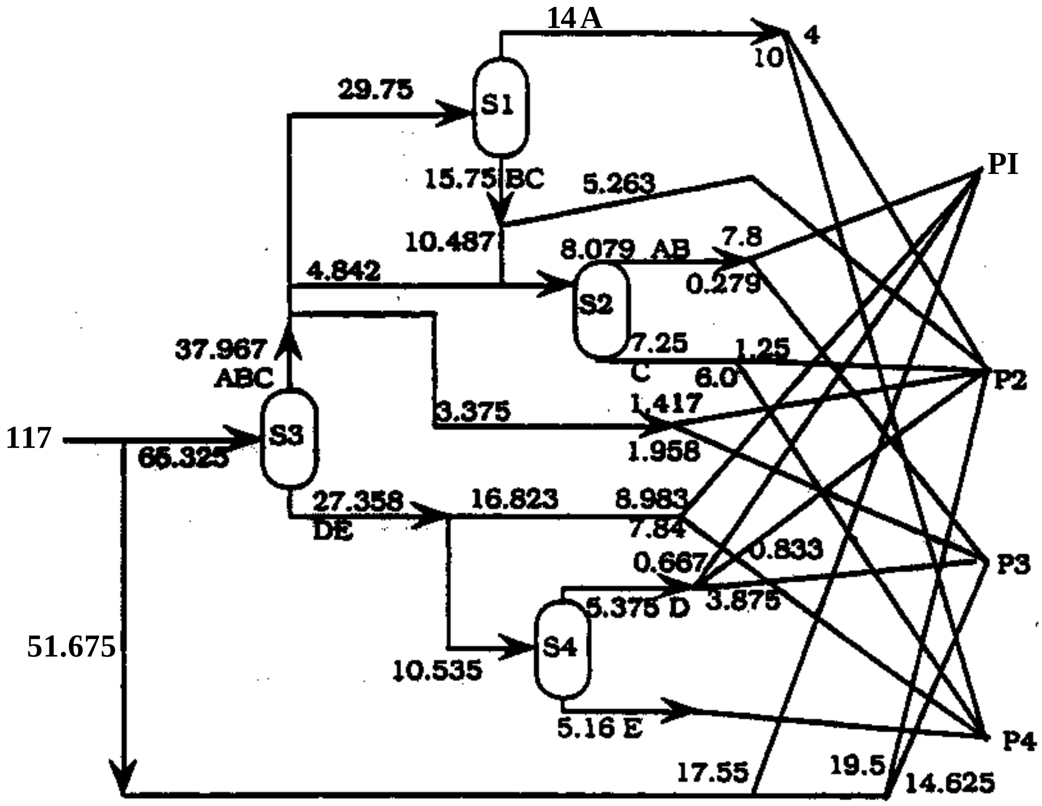


Figure 20. Solution of example 9.

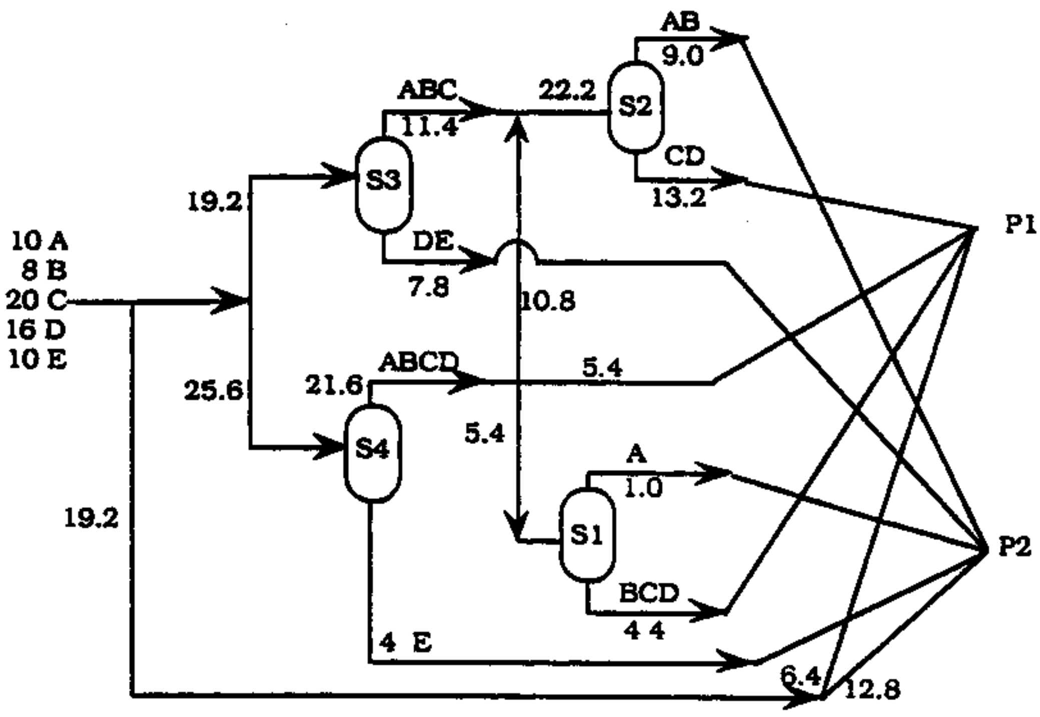


Figure 21. Solution of example 10.

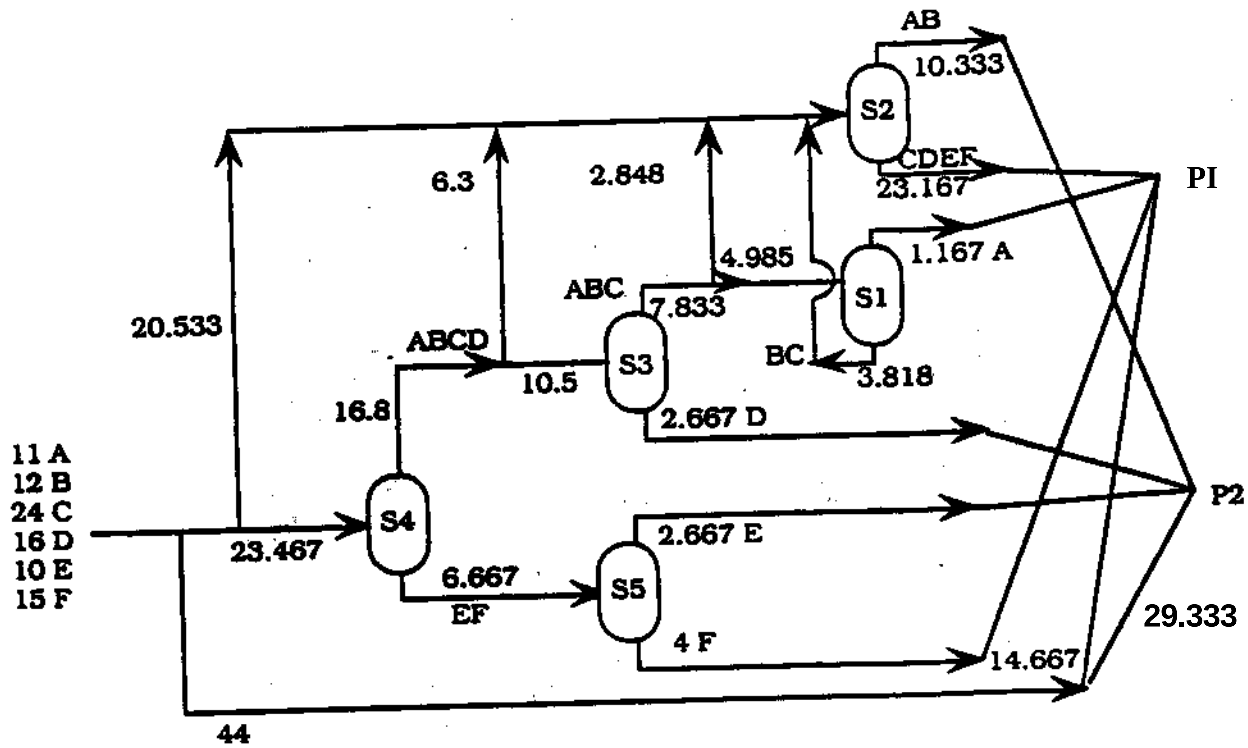


Figure 22. Solution of example 11.

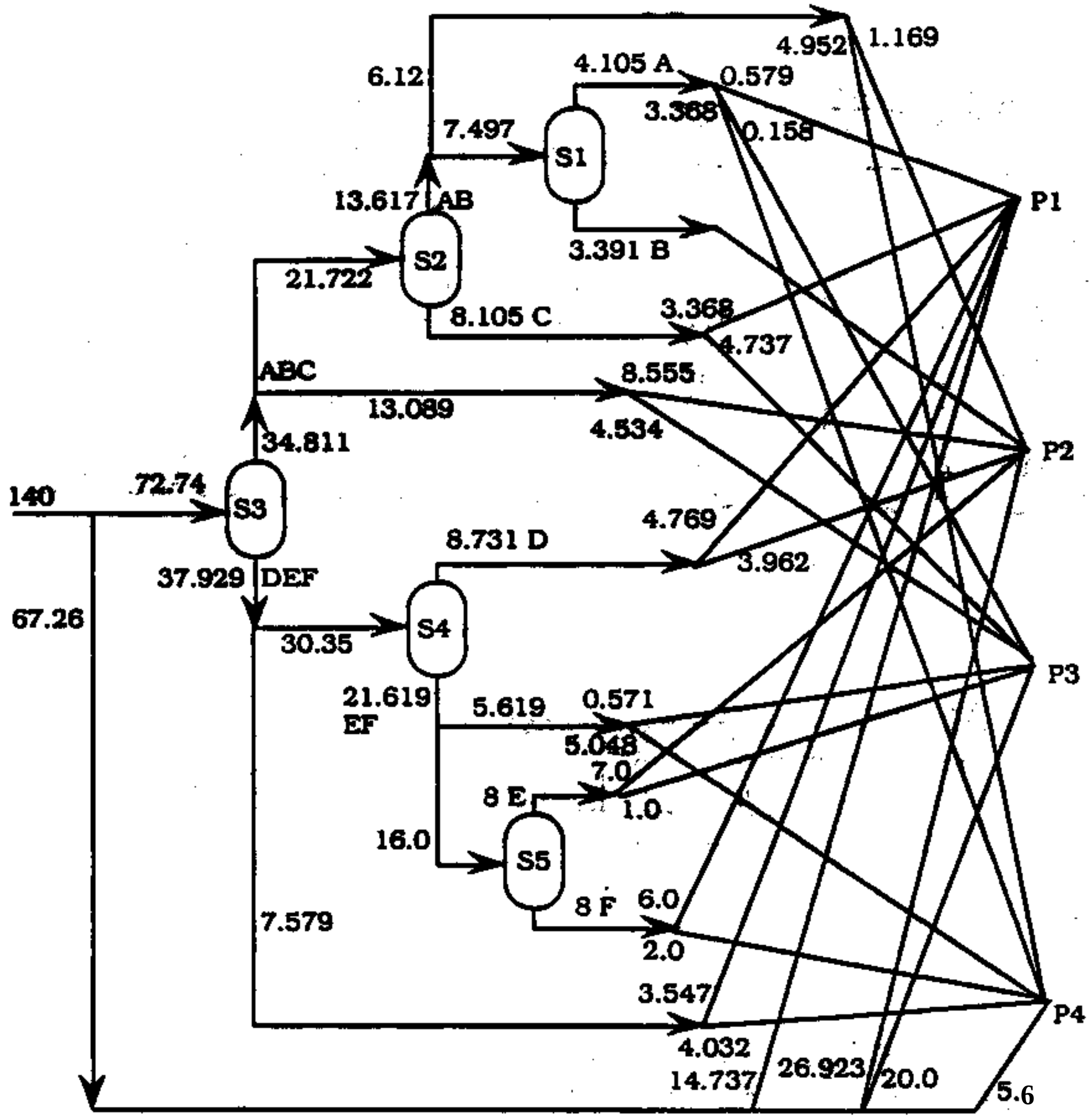


Figure 23. Solution of example 12.

Table 1. Computational results

	Comp.	>rocL	Var.	Lower bound	Initial gap	Global solution	Nodes	LP time	NLP time
Example 1	---	...	29	-500	20	-400	3	0.05	0.1
Example 2	...	...	35	-513.22	0.3	-511.87	3	0.26	0.3
Example 3			113	138.18	0.4	138.7	1	0.34	0.4
Example 4	3	2	65	1.8639	0.0	1.8639	1	0.13	—
Example 5	3	2	65	16	0.0	16	1	0.13	—
Example 6	4	2	107	54.25	2.3	55.5	3	0.97	0.4
Example 7	4	2	107	32.7	0.0	32.7	1	0.17	—
Example 8	4	3	125	26.76	0.1	26.79	1	0.23	0.3
Example 9	5	4	281	85.16	0.5	85.65	1	3.08	2.8
Example 10	5	2	225	156.56	12.4	159.48	5	2.59	2.3
Example 11	6	2	350	173	3.5	179.11	5	9.98	8.8
Example 12	6	4	430	362	14.8	388	33	19.8	13.2

Table 2. Data for example 6.

Component	A	B	C	D	Total
Product 1	5	10	4	10	29
Product 2	10	10	6	5	31
Feed	15	20	10	i5	60

Table 3. Data for example 7.

Component	A	B	C	D	Total
Product 1	7.5	10		10	31.5
Product 2	7.5	10	6	5	28.5
Feed	15	20	10	15	60

Table 4. Data for example 8.

Component	A	B	C	D	Total
Product 1	2	3	1	3	9
Product 2	1	4	1	5	11
Product 3	3	1	3	1	8
Feed	6	8	5	9	28

Table 5. Data for example 9.

Component	A	B	C	D	E	Total
Product 1	7	8	3	9	8	35
Product 2	10	3	5	5	4	27
Product 3	5	5	6	7	3	26
Product 4	10	0	6	4	9	29
Feed	32	16	20	25	24	117

Table 6, Data for example 10.

Component	A	B	C	D	E	Total
Product 1	2	2.4	16	8	1	29.4
Product 2	8	5.6	4	8	9	34.6
Feed	10	8	20	16	10	64

Table 7. Data for example 11.

Component	A	B	C	D	E	F	Total
Product 1	3	2	16	8	4	10	43
Product 2	8	10	8	8	6	5	4S
Feed	11	12	24	16	10	15	88

Table 8. Data for example 12.

Component	A	B	C	D	E	F	Total
Product 1	3	2	6	8	4	10	33
Product 2	8	10	8	8	6	5	45
Product 3	5	4	10	3	11	4	37
Product 4	7	3	1	2	5	7	25
Feed	23	19	25	21	26	26	140

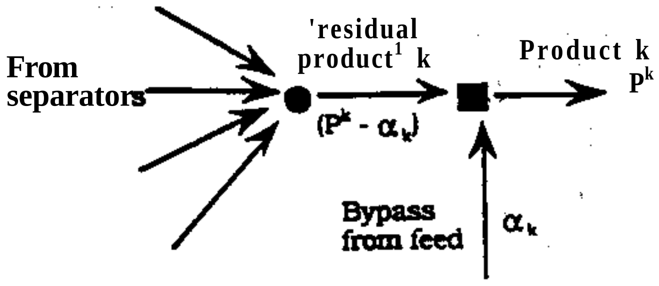


Figure 10. Definition of'residual product'.

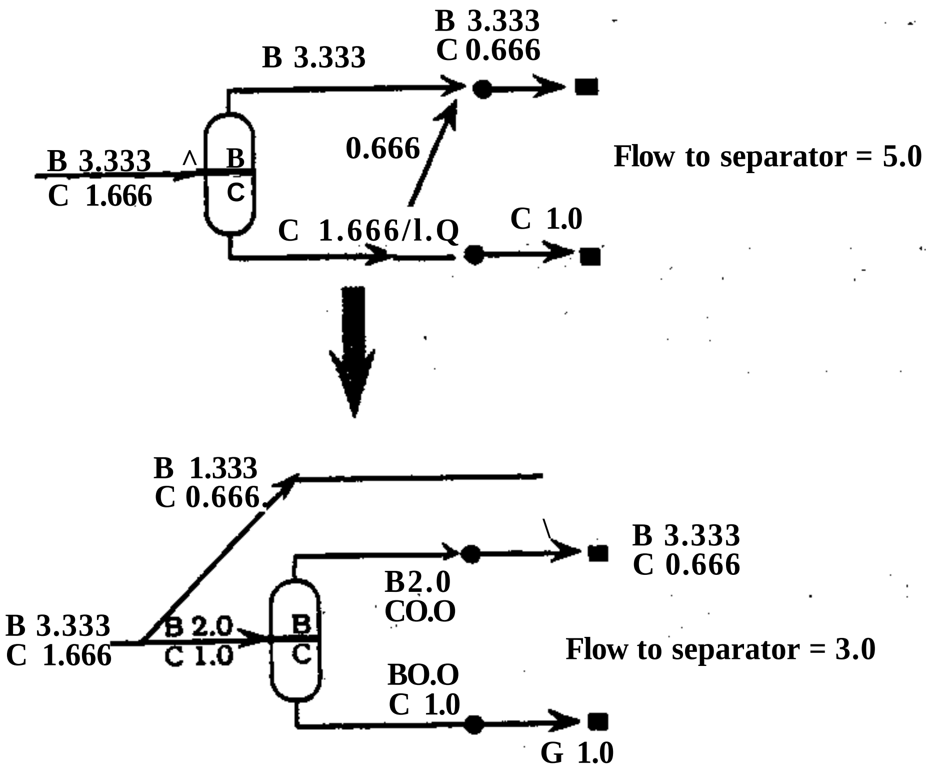


Figure 11. Example of solution without and with a zero component flow in 'residual product'.

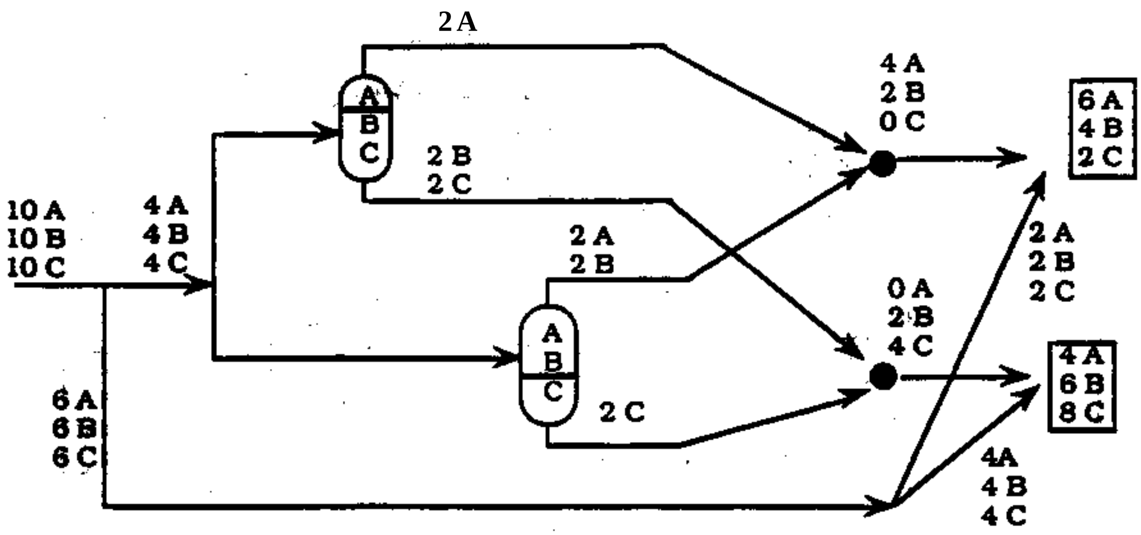


Figure 16. Global optimum solution of example 5.

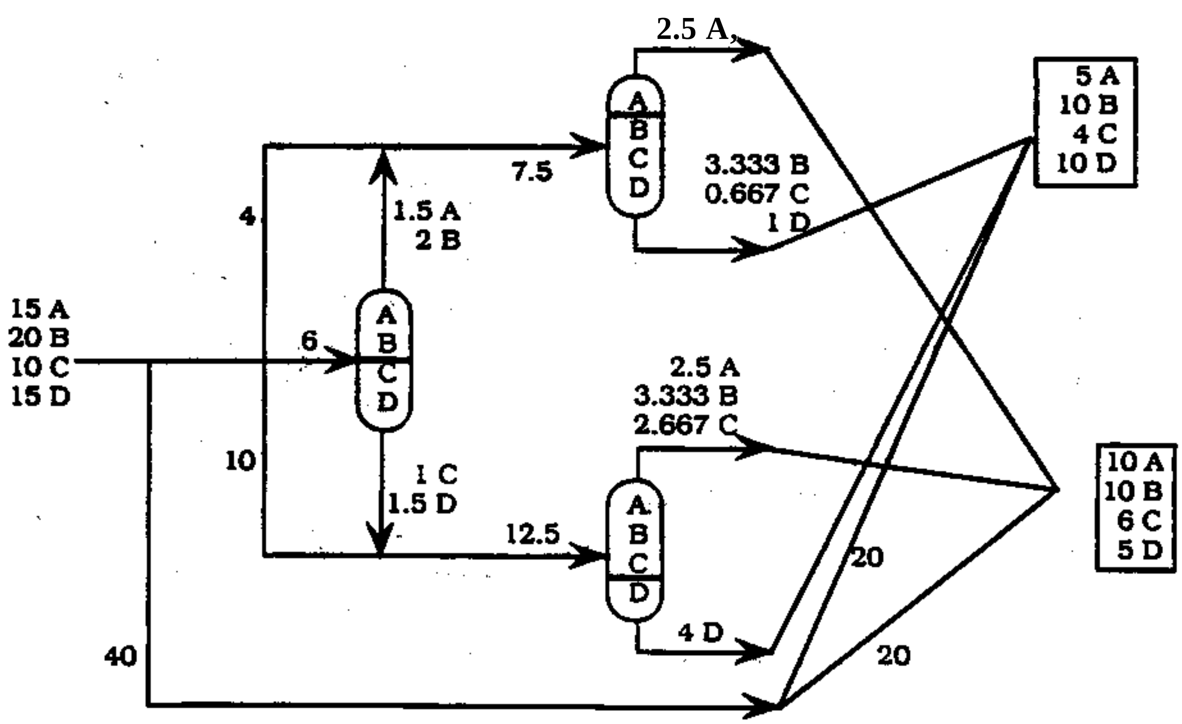


Figure 17. Solution of example 6.

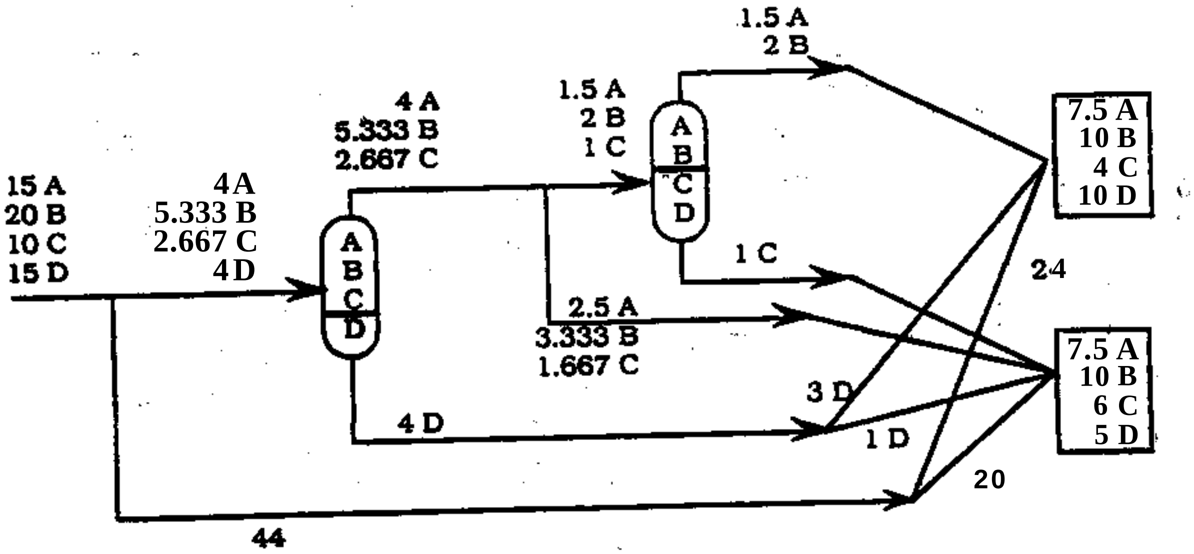


Figure 18. Solution of example 7.

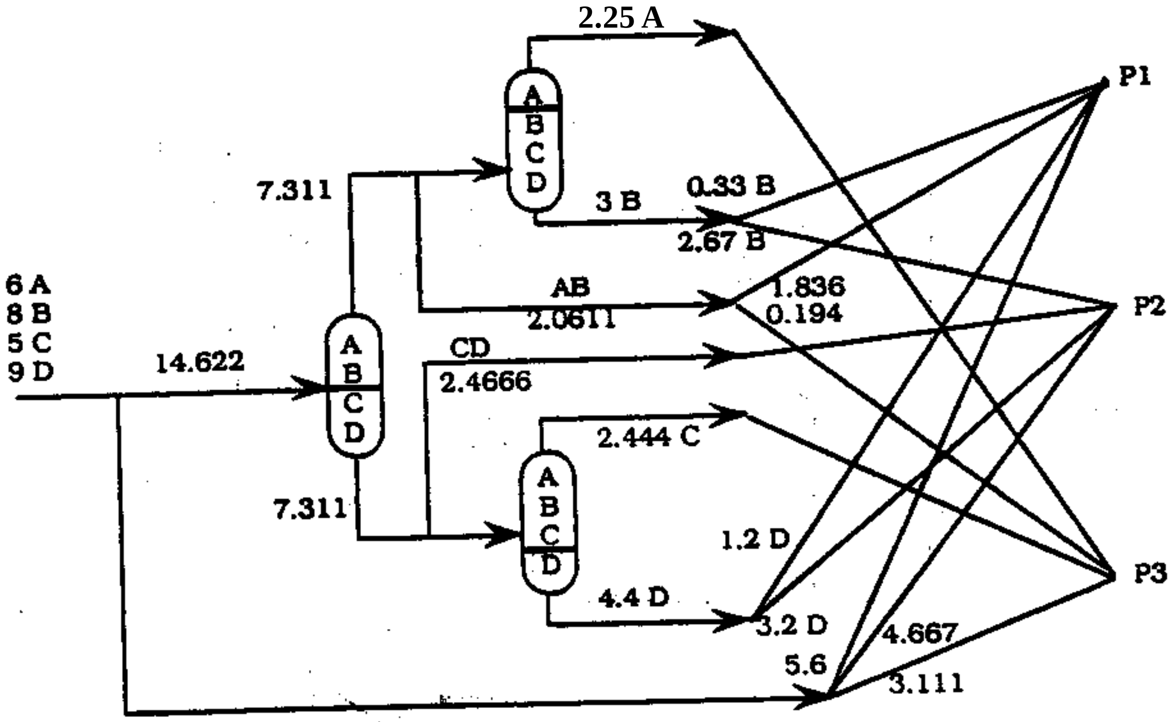


Figure 19. SoluUon of example 8.

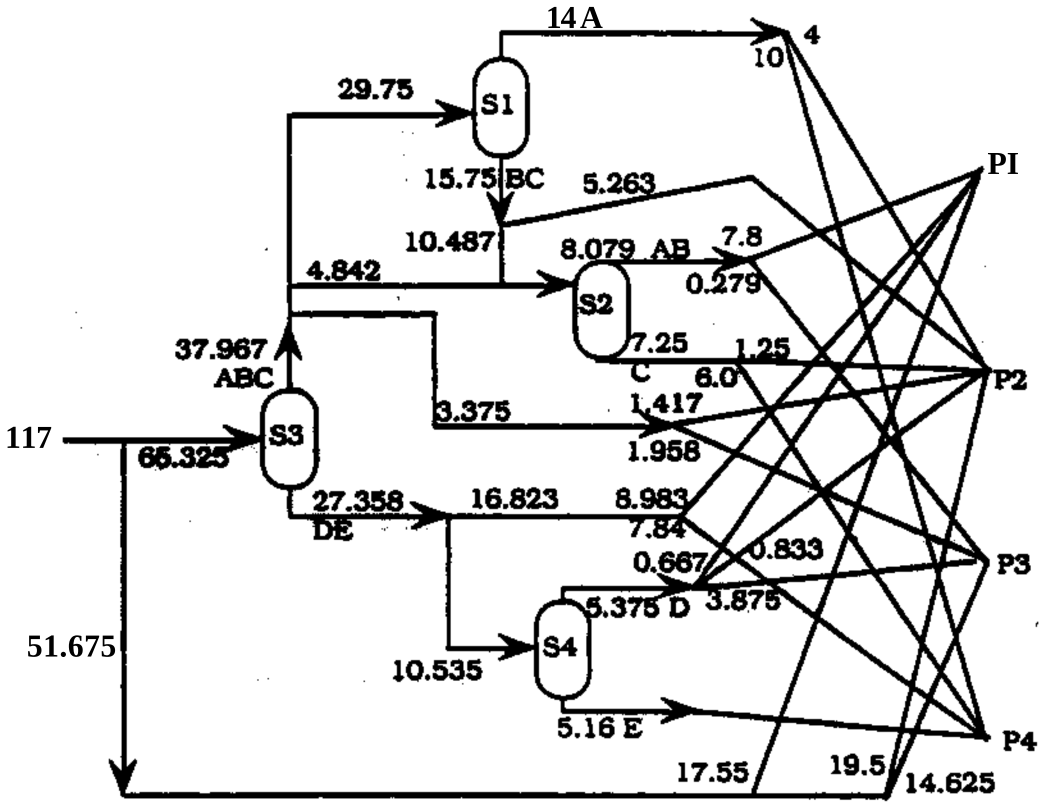


Figure 20. Solution of example 9.

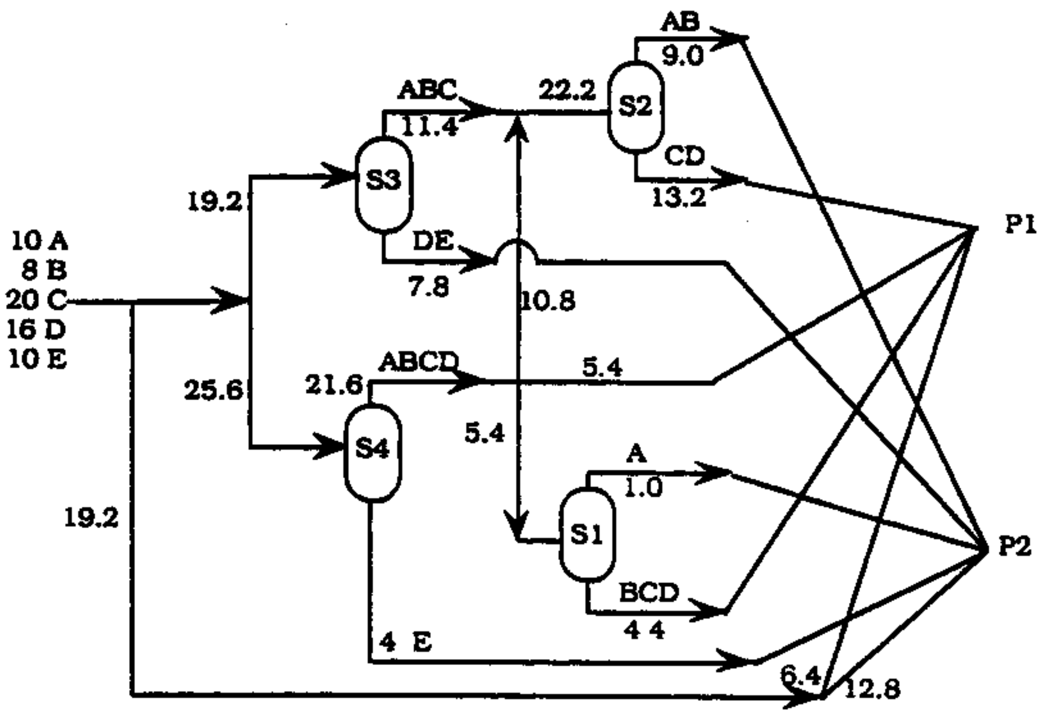


Figure 21. Solution of example 10.

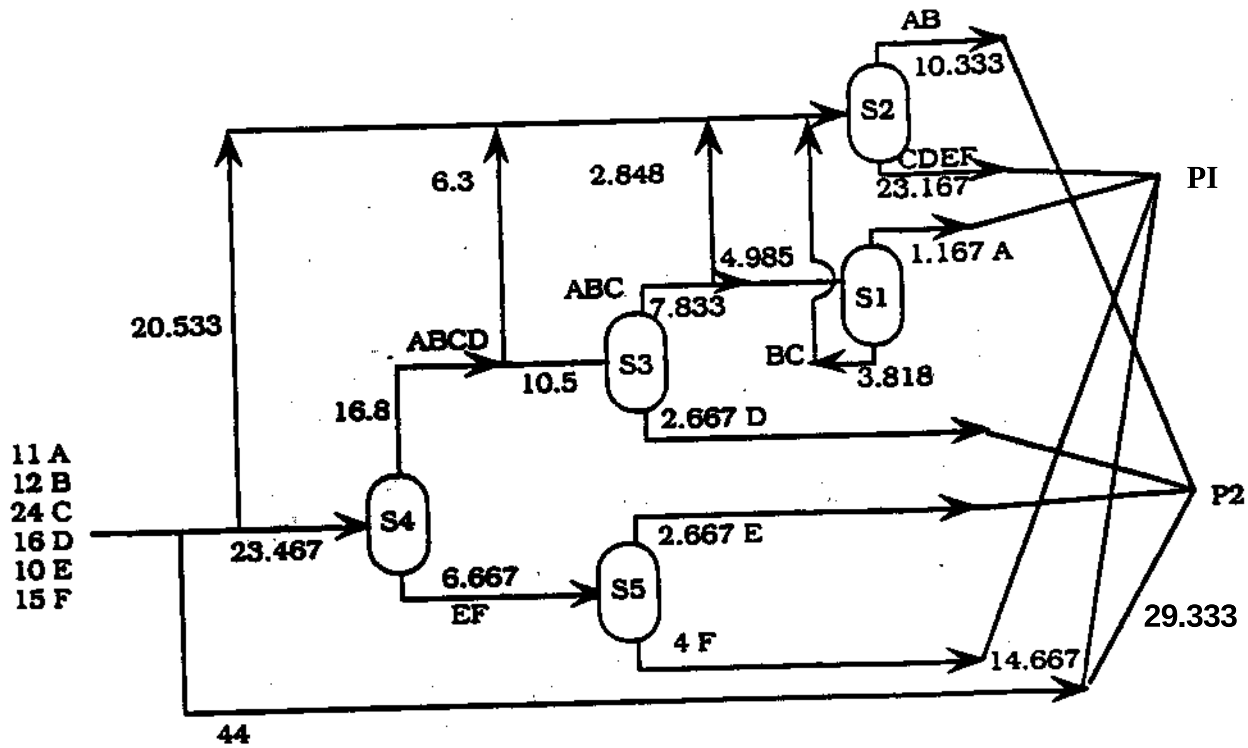


Figure 22. Solution of example 11.

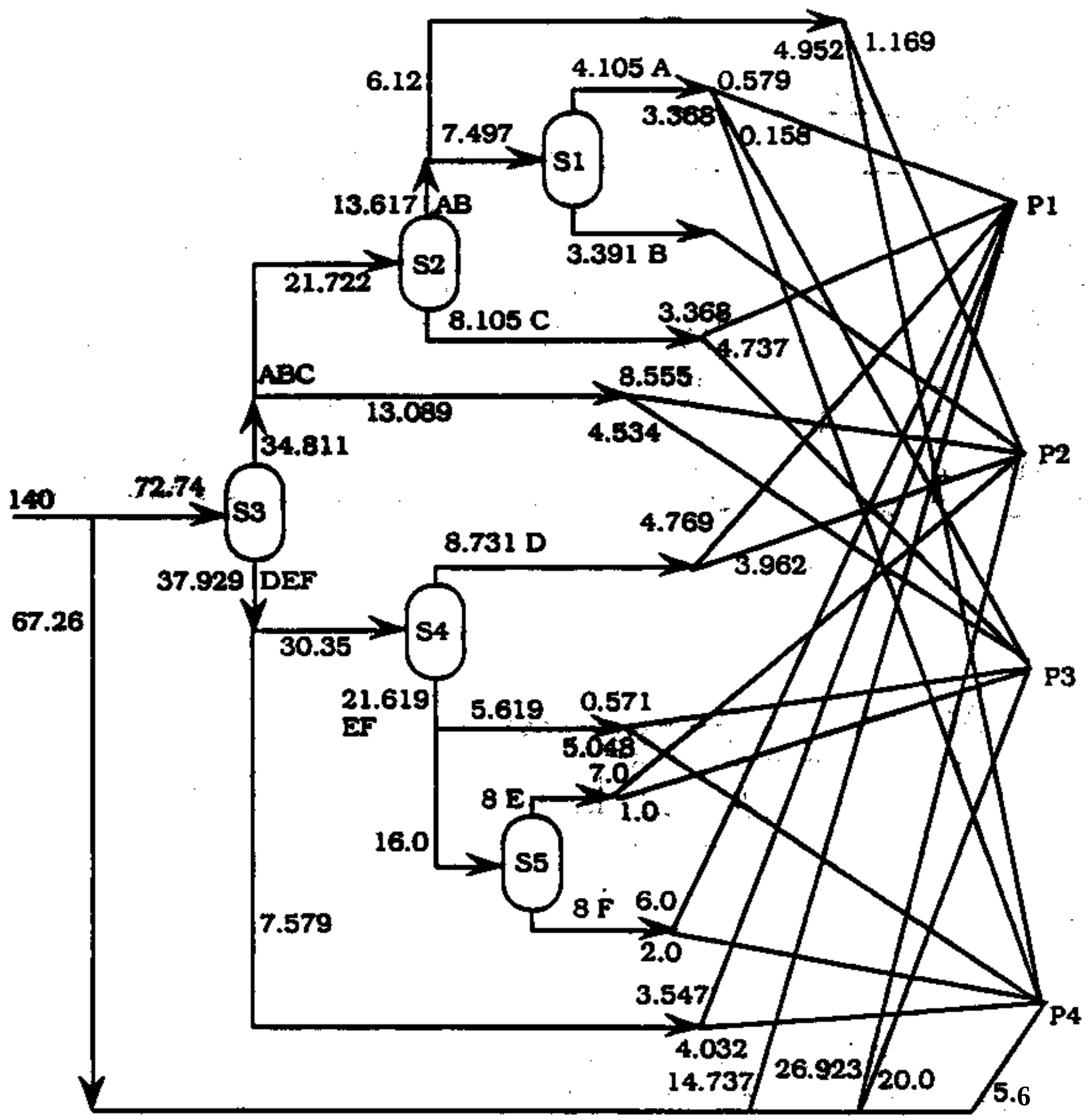


Figure 23. Solution of example 12.

Table 1. Computational results

	Comp.	Proc.	Var.	Lower bound	Initial gap	Global solution	Nodes	LP time	NLP time
Example 1	---	...	29	-500	20	-400	3	0.05	0.1
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Example 3			113	138.18	0.4	138.7	1	0.34	0.4
Example 4	3	2	65	1.8639	0.0	1.8639	1	0.13	—
Example 5	3	2	65	16	0.0	16	1	0.13	—
Example 6	4	2	107	54.25	2.3	55.5	3	0.97	0.4
Example 7	4	2	107	32.7	0.0	32.7	1	0.17	—
Example 8	4	3	125	26.76	0.1	26.79	1	0.23	0.3
Example 9	5	4	281	85.16	0.5	85.65	1	3.08	2.8
Example 10	5	2	225	156.56	12.4	159.48	5	2.59	2.3
Example 11	6	2	350	173	3.5	179.11	5	9.98	8.8
Example 12	6	4	430	362	14.8	388	33	19.8	13.2

Table 2. Data for example 6.

Component	A	B	C	D	Total
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Product 2	10	10	6	5	31
Feed	15	20	10	15	60

Table 3. Data for example 7.

Component	A	B	C	D	Total
Product 1	7.5	10		10	31.5
Product 2	7.5	10	6	5	28.5
Feed	15	20	10	15	60

Table 4. Data for example 8.

Component	A	B	C	D	Total
Product 1	2	3	1	3	9
Product 2	1	4	1	5	11
Product 3	3	1	3	1	8
Feed	6	8	5	9	28

Table 5. Data for example 9.

Component	A	B	C	D	E	Total
Product 1	7	8	3	9	8	35
Product 2	10	3	5	5	4	27
Product 3	5	5	6	7	3	26
Product 4	10	0	6	4	9	29
Feed	32	16	20	25	24	117

Table 6, Data for example 10.

Component	A	B	C	D	E	Total
Product 1	2	2.4	16	8	1	29.4
Product 2	8	5.6	4	8	9	34.6
Feed	10	8	20	16	10	64

Table 7. Data for example 11.

Component	A	B	C	D	E	F	Total
Product 1	3	2	16	8	4	10	43
Product 2	8	10	8	8	6	5	45
Feed	11	12	24	16	10	15	88

Table 8. Data for example 12.

Component	A	B	C	D	E	F	Total
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Product 2	8	10	8	8	6	5	45
Product 3	5	4	10	3	11	4	37
Product 4	7	3	1	2	5	7	25
Feed	23	19	25	21	26	26	140

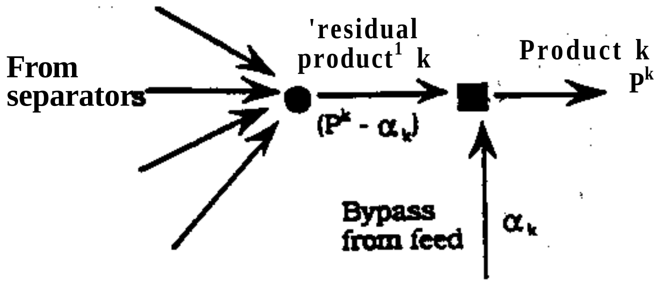

Figure 10. Definition of 'residual product'.

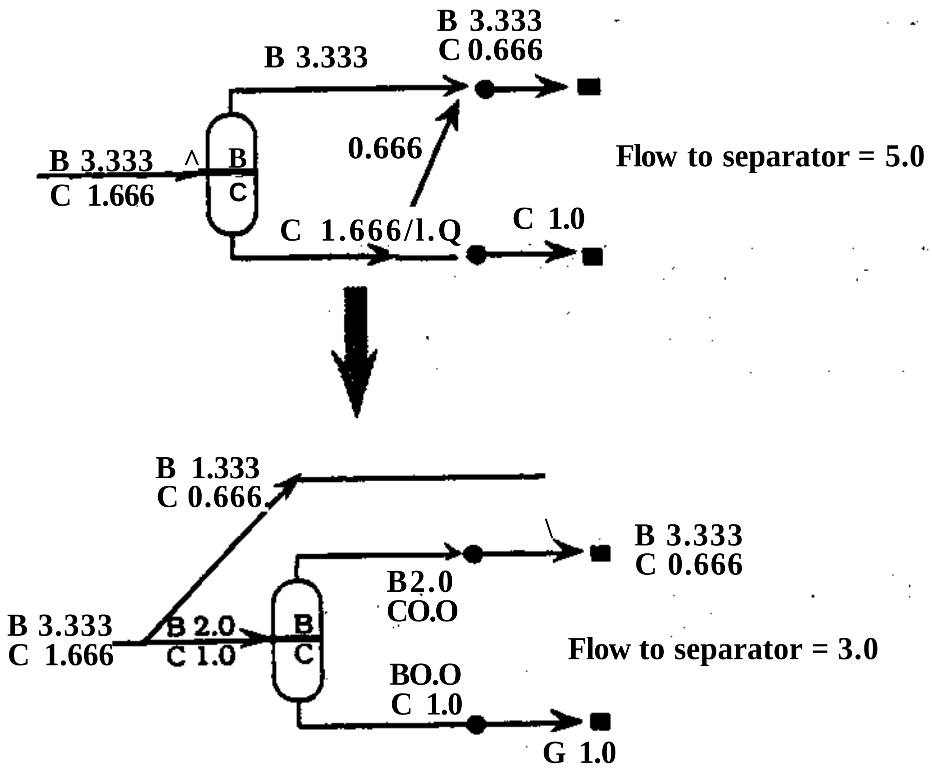

Figure 11. Example of solution without and with a zero component flow in 'residual product'.