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Minimization of cost of recycling in chemical processes

Gautham Parthasarathy*

Department of Chemical Engineering, Auburn University, 230 Ross Hall, Auburn, AL 36849, USA

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Abstract

This paper considers an important aspect of every chemical process. Most chemical processes have streams being recycled back to different process units. These streams have to meet certain process requirements such as flow rate and composition. Besides these material balance constraints, their relative costs have to be considered. There is a need to determine the *global* cheapest mixing and recycling scheme for a given process. A generic non-linear program formulation is given and a solution algorithm consisting of two different levels is proposed. The outer level consists of iterations among different variables while the inner level consists of solution of a linear program. In the case of lower number of sources and sinks, this paper introduces application of the simplex algorithm to derive frameworks wherein, the optimal flow rates of the different sources can be obtained as functions of relative costs and compositions. These frameworks make it possible to determine the cheapest recycling scheme for all available streams. Three cases with progressively increasing complexity are considered. These cases cover a large number of potential industrial applications. This methodology is equally applicable to liquid, solid and gaseous streams. It can handle streams coming from unit operations such as crystallization, condensation and adsorption. It requires basic information on flow rate and compositions of all the sources and sinks being considered and the unit costs of the various sources involved. It is easy to apply and is rigorous under the conditions listed for its use. Two different case studies are considered and the application of the proposed solution algorithm is demonstrated.

Keywords: Recycling; Cost minimization; Global solution algorithm; Simplex; Mass integration; Crystallization; Condensation; Adsorption; Penicillin; Urea-adduct process

1. Introduction

A typical chemical process consists of a series of processing steps in various units from initial reactants to final products. The sequence of steps to be followed is determined by the specific process under consideration. However, most chemical processes have one feature in common: there are a number of streams being recycled back to different process units. These streams typically have to meet certain process requirements (such as flow rate and composition) of the unit to which they are being recycled. The proportions of various streams that are mixed and/or recycled back are usually guided by process demands. These process requirements can be met by a large number of permutations and combinations among the streams available for potential recycle. This paper deals with developing a systematic framework to determine the cheapest possible recycling scheme that would satisfy all process requirements. This work can be applied to a wide range of process operations. It can be used to determine the amounts of fresh materials (e.g. solvents, and reagents) to be added along with recycled streams as part of make-up to a process. It can also be used to generate the cheapest recycle and allocation strategies for condensation, crystallization and adsorption systems.

2. Previous work

For any given process, sources and sinks can be defined. A source is any stream in the process carrying a species of interest while a sink is any unit in the process (such as an absorption or distillation column) that is capable of handling the source. A source may consist of fresh species (i.e. a fresh source) or may have been generated in the process (i.e. a process source). The fresh sources are typically obtained externally (either bought or imported from another process) and is composed of either one pure species or a mixture of pure species. Their cost is dependent primarily on the market value of the pure species present in them. On the other hand, process sources are typically composed of more than one species and their cost depends on the manner in which they were created. Due to the limitations of the process generating them, there may exist upper bounds on the flow rates of the process sources.

* Tel.: +1-334-844-2061; fax: +1-334-844-2063.

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Nomenclature

C_m	Unit cost of *m*th source (US$/kg)
j	index for each sink
k	index for each species
L_m	Total flow rate of *m*th source (kg/s)
L_j^{sink}	Total inlet flow rate of *j*th sink (kg/s)
$l_{m,j}$	Individual flow rate from *m*th source to *j*th sink (kg/s)
m	index for each source
mincost	variable storing optimal minimum cost solution value
$N1_j$	number of iterations for total sink inlet flow rate of *j*th sink
$N2_{j,k}$	number of iterations for composition of *k*th species in *j*th sink
$N3_m$	number of iterations for flow rate of *m*th source
$N4_{m,k}$	number of iterations for composition of *k*th species of *m*th source
N_{large}	arbitrarily selected large number
N_{sink}	total number of sinks
N_{source}	total number of sources
N_{species}	total number of species
t_j	iteration index corresponding to total sink inlet flow rate of *j*th sink
$u_{j,k}$	iteration index corresponding to composition of *k*th species in *j*th sink
v_m	iteration index corresponding to flow rate of *m*th source
$w_{m,k}$	iteration index corresponding to composition of *k*th species of *m*th source
$x_{m,k}$	composition of *k*th species in *m*th source
$z_{j,k}^{\text{sink}}$	Inlet composition of *k*th species for *j*th sink

Subscripts

l	refers to lower bound
lower	refers to lower bound
sink	refers to sink
source	refers to source
species	refers to species
u	refers to upper bound
upper	refers to upper bound

Superscripts

sink	refers to sink
T	refers to sink
u	refers to upper bound

Greek Letters

α	relational cost parameter
α_m	known upper bound on flow rate of *m*th process source (kg/s)
β_j	known inlet flow rate of *j*th sink (kg/s)
$\chi_{j,k}$	known inlet mass load of *k*th species to *j*th sink (kg/s)
θ	variable calculated as part of respective simplex algorithm

The chemical process industry is lowering costs and improving productivity via increased process integration. Mass integration is a critical element of this effort. Early work in this area focused on the synthesis of separation networks that employed mass separating agents or MSAs. El-Halwagi and Manousiouthakis [1] introduced the concept of mass exchange networks or MENs. Several forms of the MEN problem were later defined and addressed. These include optimization based techniques for handling single [2], multiple components [3,4] and simultaneous synthesis of mass exchange and regeneration networks [5,6]. Papalexandri and Pistikopoulos [7] developed a structural approach to the synthesis of MENs. Kiperstok and Sharratt [8] optimized MENs for the removal of pollutants. Another class of separation networks is induced by the use of energy separating agents or ESAs. Examples of these energy induced separation networks include crystallization [9–11], distillation [12–16], membrane separations [17,18], evaporation, drying and condensation [19–21]. Separation networks constitute only a subset of available mass integration strategies. In most cases, the recovered species can and should be recycled/reused in the process. Hence, separation has to be integrated with decisions on how much of each species to recover and how the recovered species will be used. Thus, we need to invoke the more general concept of mass integration, which is a holistic approach that deals with the optimum generation, separation, and routing of streams and species [22–24]. El-Halwagi et al. [22] came up with the concept of waste interception and allocation networks that shifted the entire focus from end of pipe treatment to in plant treatment. A particularly useful framework for integration, allocation, generation and separation of streams and species is given in Fig. 1.

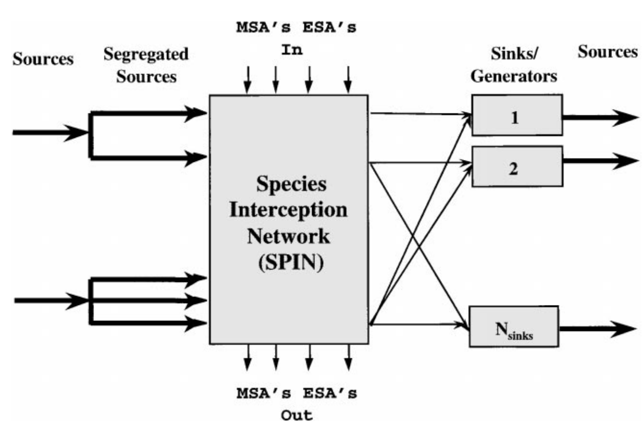

Fig. 1. Mass integration framework [22–24].

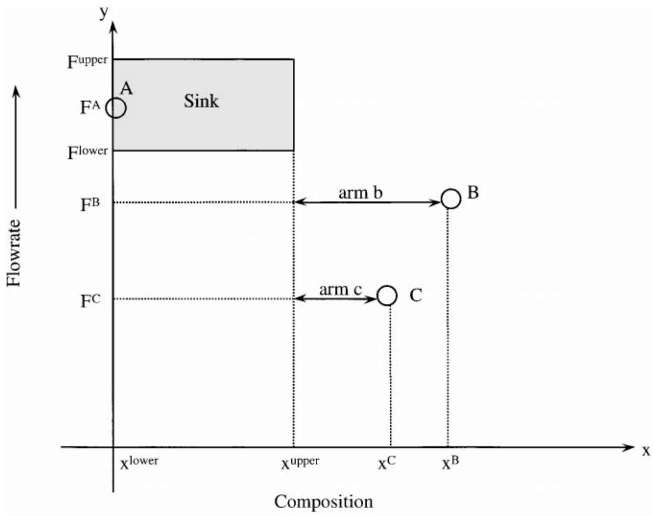

Fig. 2. Mixing and recycling strategies for dilute single component systems.

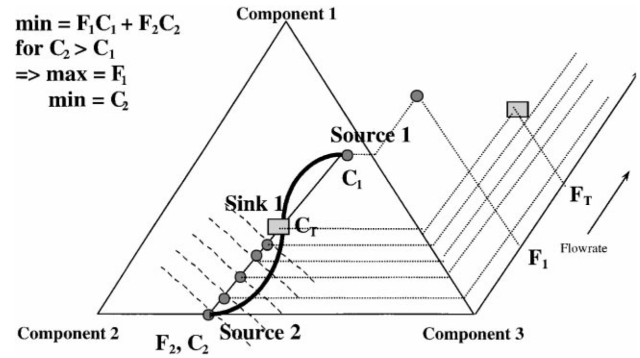

Fig. 3. Mixing and recycling rule for ternary system with single source and sink.

The mass integration approach uses different strategies to achieve its targets. These include segregation (i.e. avoiding stream mixing), mixing and recycling (i.e. sending a particular source to a specified sink). The mixing and recycling strategies adopted are not arbitrary but are generated by the application of rigorous rules. For dilute systems, El-Halwagi [25] describes graphical mixing rules for the case of a single pollutant present in aqueous sources.

Let us assume we have two pollutant laden sources, B and C, and one process sink, which can accept these sources. The sink will have upper and lower bounds on the composition (x^{upper} and x^{lower}) and flow rates (F^{upper} and F^{lower}) of the sources that it can accept. As shown in Fig. 2, the sink is represented as a box while the sources are represented as points. This diagram is called as the source sink diagram. Fresh water is represented on the *y*-axis as its pollutant composition is assumed to be zero. Currently, the fresh water is being supplied to the sink via source A and it is desired to lower fresh water requirement by mixing and recycling sources B and/or C to the sink. In this case, the lever arm rule for mixing is applied. The source with the least arm should be recycled so as to minimize the fresh water requirement. As arm ‘c’ is smaller than arm ‘b’ in Fig. 2, the strategy to minimize fresh water is to mix source C with fresh water, followed by recycle to the sink. For dilute streams with multiple components, similar analysis can be carried out by assuming that each species behaves independent of the other species. As the species being tracked is present in very low concentrations, the cost of mixing and recycling is primarily contributed by the fresh water requirement. Therefore, the cost of mixing and recycling is minimized at minimum fresh water usage in the process.

However, in the case of sources with high concentrations of the species of interest, the cost of mixing and recycling will be a function of all species present. Some papers have addressed the problem of obtaining minimum cost solutions to the condensation and allocation of multiple volatile organic compounds [26]. Graphical solutions were developed for the case of four component systems (e.g. an inert carrier and three VOCs). A triangular source sink diagram was introduced with the triangle acting as the composition plane. Mixing rules were developed to determine allocation of various sources to different sinks such that the overall cost is minimized. An example is given in Fig. 3. This rule determines the location of a source for mixing and recycling given a source and sink with known locations on the composition triangle.

Source 1, at a composition C_1 and flow rate F_1, is to be mixed with source 2 (whose location in the composition triangle is unknown and has to be determined) such that the resultant stream satisfies the requirements for sink 1 with compositions C_T and flow rate F_T. From the lever arm rule for mixing, source 2 will lie on a straight line joining sink 1 and source 1 but on the other side of sink 1. The cost objective function is given by $F_1C_1+F_2C_2$, which is minimized when, for $C_2>C_1$, we maximize F_1. For this condition to be satisfied, the lever arm rule dictates that the arm corresponding to source 2 should be maximized. So, source 2 will lie on a straight line joining sink 1 and source 1 but as far away as possible on the other side of sink 1.

However, a drawback of all these rules is that they are valid under specific conditions. There is a need for a method to determine the minimum cost of mixing for a generic process. The graphical approach is also limited to cases with dilute streams or specific cases of concentrated systems. As described previously for the VOCs, all the sinks and sources have fixed compositions and flow rates. However, in industrial applications, the flow rates and compositions of both the sources and sinks can fluctuate. Therefore, it is more accurate to define an operating range for each source and sink. In this case, it would be difficult to come up with a generic rule to determine minimum cost of recycling, as there are a multitude of possible flow rates and compositions for each source and sink being considered.

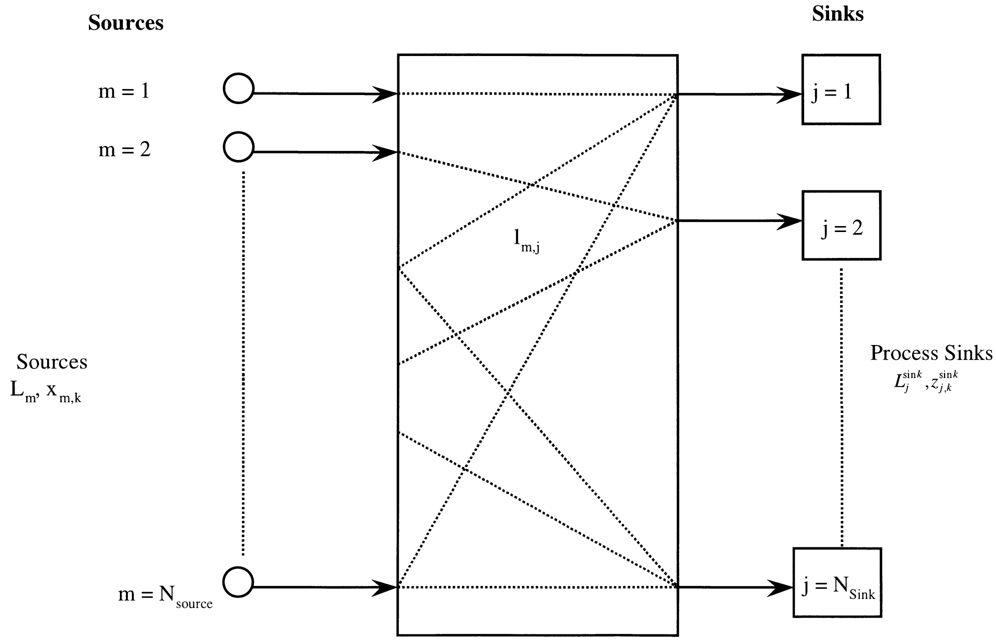

Fig. 4. Optimal mixing and allocation representation.

3. Problem definition

The problem may be given as

$$\min = \sum_{m=1}^{N_{\text{source}}} L_m C_m \tag{1}$$

For each source m, L_m and C_m refer to the total source flow rate and unit cost respectively while $x_{m,k}$ refers to the source composition, where k is the index for each species present. The cost of each fresh source is the cost of fresh species used to make the source. The cost of each process source is the cost involved in synthesizing the source (e.g. if the source was obtained via crystallization then the total cost of crystallization divided by the total amount of the source crystallized out would be the unit cost of the process source). For each sink j, L_j^{sink} refers to the total sink inlet flow rate, $z_{j,k}^{\text{sink}}$ refers to the sink inlet composition, where k is the index for each species present. Also, $l_{m,j}$ refers to the individual flow going from source m to sink j.

The following constraints would describe the mixing and allocation problem and incorporates all the segregation, mixing and recycle constraints (refer Fig. 4).

Source material balance:

$$L_m = \sum_{j=1}^{N_{\text{sink}}} l_{m,j}, \quad m = 1, 2, \ldots, N_{\text{source}} \tag{2}$$

L_m refers to both process and fresh sources. For process sources, there may exist upper bounds on the total flow rate available for recycle. For fresh sources, there is no upper bound as these are typically purchased externally.

Sink material balance:

$$L_j^{\text{sink}} = \sum_{m=1}^{N_{\text{source}}} l_{m,j}, \quad j = 1, 2, \ldots, N_{\text{sink}} \tag{3}$$

Sink inlet composition balance:

$$L_j^{\text{sink}} z_{j,k}^{\text{sink}} = \sum_{m=1}^{N_{\text{source}}} l_{m,j} x_{m,k}, \quad j = 1, 2, \ldots, N_{\text{sink}}, \quad k = 1, 2, \ldots, N_{\text{species}} \tag{4}$$

Source limits:

$$x_{m,k,\text{lower}} \leq x_{m,k} \leq x_{m,k,\text{upper}}, \quad L_{m,\text{lower}} \leq L_m \leq L_{m,\text{upper}}, \quad m = 1, 2, \ldots, N_{\text{source}}, \quad k = 1, 2, \ldots, N_{\text{species}} \tag{5}$$

Sink inlet limits:

$$z_{j,k,\text{lower}}^{\text{sink}} \leq z_{j,k}^{\text{sink}} \leq z_{j,k,\text{upper}}^{\text{sink}}, \quad L_{j,\text{lower}}^{\text{sink}} \leq L_j^{\text{sink}} \leq L_{j,\text{upper}}^{\text{sink}}, \quad j = 1, 2, \ldots, N_{\text{sink}}, \quad k = 1, 2, \ldots, N_{\text{species}} \tag{6}$$

4. Design challenges

As the problem requires non-linear optimization, a global solution is not guaranteed. There exist a large number of potential flow rates and compositions for each of the process sources and sinks under consideration. This implies a large number of combinations among various streams to meet the requirements of the different process sinks. The optimal solution is a function of the material balance and relative costs of the different sources present in the process. There may also exist multiple components in the system that increases the complexity of the problem. These aspects combine to make the above-defined problem challenging to solve.

5. Solution strategy

This non-linear optimization problem can be solved via the proposed two level solution strategy. It may be noted that the non-linear program reduces to a linear problem in the special case of fixed compositions and flow rates of all sources and sinks being considered.

$$\min = \sum_{m=1}^{N_{\text{source}}} L_m C_m \tag{7}$$

subject to

$$\sum_{j=1}^{N_{\text{sink}}} l_{m,j} = L_m \leq \alpha_m \tag{8}$$

For process sources, the upper bound on flow rate is known and represented by α_m, while for fresh sources, Eq. (8) is given as

$$\sum_{j=1}^{N_{\text{sink}}} l_{m,j} = L_m \tag{8}$$

$$\sum_{m=1}^{N_{\text{source}}} l_{m,j} = L_j^{\text{sink}} = \beta_j \tag{9}$$

$$\sum_{m=1}^{N_{\text{source}}} l_{m,j} x_{m,k} = L_j^{\text{sink}} z_{j,k}^{\text{sink}} = \chi_{j,k} \tag{10}$$

The above formulation is a linear program with all linear constraints for known values of α_m, β_j, $\chi_{j,k}$ and compositions. This program can be evaluated globally.

As part of the global solution strategy, the given problem is divided into two levels. The first outer level considers all possible values of the various sources and sink compositions and flow rates while the second inner level gives the optimal mixing ratio of available sources to satisfy the requirements of all sinks at minimum cost. The inner level involves the solution of a linear program to come up with optimal recycling strategies. Once these strategies have been formulated, the outside iteration is defined and the solution algorithm is complete.

Given subsequently is the solution methodology to be followed to come up with a global solution to this non-linear optimization problem.

1. The following increments are defined.

$$\Delta L_j^{\text{sink}} = \frac{L_{j,\text{upper}}^{\text{sink}} - L_{j,\text{lower}}^{\text{sink}}}{N1_j}, \quad j = 1, 2, \ldots, N_{\text{sink}} \tag{11}$$

$$\Delta z_{j,k}^{\text{sink}} = \frac{z_{j,k,\text{upper}}^{\text{sink}} - z_{j,k,\text{lower}}^{\text{sink}}}{N2_{j,k}}, \quad j = 1, 2, \ldots, N_{\text{sink}},\ k = 1, 2, \ldots, N_{\text{species}} - 1 \tag{12}$$

$$\Delta L_m = \frac{L_{m,\text{upper}} - L_{m,\text{lower}}}{N3_m}, \quad m = 1, 2, \ldots, N_{\text{source}} \tag{13}$$

$$\Delta x_{m,k} = \frac{x_{m,k,\text{upper}} - x_{m,k,\text{lower}}}{N4_{m,k}}, \quad m = 1, 2, \ldots\ldots\ldots, N_{\text{source}},\ k = 1, 2, \ldots\ldots\ldots, N_{\text{species}} - 1 \tag{14}$$

2. We define four sets of indices namely, t_j, $u_{j,k}$, v_m, and $w_{m,k}$ corresponding to L_j^{sink}, $z_{j,k}^{\text{sink}}$, L_m and $x_{m,k}$, respectively. The variable mincost is defined to contain the minimum cost global solution and is set initially to an arbitrarily high value, N_{large}. All indices are initially set to zero.
3. The outer level of the solution procedure consists of iterations among all permissible values of the variables L_j^{sink}, $z_{j,k}^{\text{sink}}$, L_m and $x_{m,k}$ The inner level consists of a linear problem that can be solved globally. For each of the above variables

$$L_j^{\text{sink}} = L_{j,\text{upper}}^{\text{sink}} - t_j \Delta L_j^{\text{sink}}, \quad j = 1, 2, \ldots, N_{\text{sink}} \tag{15}$$

$$z_{j,k}^{\text{sink}} = z_{j,k,\text{upper}}^{\text{sink}} - u_{j,k} \Delta z_{j,k}^{\text{sink}}, \quad j = 1, 2, \ldots, N_{\text{sink}},\ k = 1, 2, \ldots, N_{\text{species}} - 1 \tag{16}$$

$$L_m = L_{m,\text{upper}} - v_m \Delta L_m, \quad m = 1, 2, \ldots, N_{\text{source}} \tag{17}$$

$$x_{m,k} = x_{m,k,\text{upper}} - w_{m,k} \Delta x_{m,k}, \quad m = 1, 2, \ldots, N_{\text{sink}},\ k = 1, 2, \ldots, N_{\text{species}} - 1 \tag{18}$$

As all the compositions should add up to 1.0

$$z_{j,N_{\text{species}}}^{\text{sink}} = 1.0 - \sum_{k=1}^{N_{\text{species}}-1} z_{j,k}^{\text{sink}}, \quad j = 1, 2, \ldots, N_{\text{sink}} \tag{19}$$

$$x_{m,N_{\text{species}}} = 1.0 - \sum_{k=1}^{N_{\text{species}}-1} x_{m,k}, \quad m = 1, 2, \ldots, N_{\text{source}} \tag{20}$$

The composition of any of the N_{species} present can be calculated by difference using the above two equations.

Each of the iterations in the outer loop is carried out till the following conditions are satisfied:

$$t_j \leq N1_j, \quad j = 1, 2, \ldots, N_{\text{sink}} \tag{21}$$

$$u_{j,k} \leq N2_{j,k}, \quad j = 1, 2, \ldots, N_{\text{sink}}, \quad k = 1, 2, \ldots, N_{\text{species}} - 1 \tag{22}$$

$$v_m \leq N3_m, \quad m = 1, 2, \ldots, N_{\text{source}} \tag{23}$$

$$w_{m,k} \leq N4_{m,k}, \quad m = 1, 2, \ldots, N_{\text{source}}, \quad k = 1, 2, \ldots, N_{\text{species}} - 1 \tag{24}$$

4. The inner level consists of calculation of minimum cost for a linear program and storing this value in the variable mincost. For the first iteration, mincost is set equal to N_{large}. For all subsequent iterations, the calculated cost is compared with the previous minimum value and the least of the two values is stored. Once all iterations are completed, the minimum cost global solution is obtained. An example of a typical solution algorithm is given in Fig. 5. This algorithm is applicable for the case of a system with two fresh species present with one process source and one process sink. There are four iteration indices, *i*, *j*, *k* and *l*, which are tracking f^{T}, x^{T}, *f* and *x*, respectively. *i*_min, *j*_min, *k*_min and *l*_min are the respective values of these iteration indices at the global optimal point. f^{T} and x^{T} refer to total inlet flow rate and composition of the process sink, while *f* and *x* refer to the flow rate and composition of the process source. f_{u}^{T}, x_{u}^{T}, f_{u} and x_{u}, and f_{l}^{T}, x_{l}^{T}, f_{l} and x_{l} refer to the respective upper and lower bounds on each of the variables. Four different increments are defined as follows:

$$Df^{\text{T}} = \frac{f_{\text{u}}^{\text{T}} - f_{\text{l}}^{\text{T}}}{N_1} \tag{25}$$

$$Dx^{\text{T}} = \frac{x_{\text{u}}^{\text{T}} - x_{\text{l}}^{\text{T}}}{N_2} \tag{26}$$

$$Df = \frac{f_{\text{u}} - f_{\text{l}}}{N_3} \tag{27}$$

$$Dx = \frac{x_{\text{u}} - x_{\text{l}}}{N_4} \tag{28}$$

N_1, N_2, N_3 and N_4 are pre-defined numbers, which can be increased to have lower increments.

The linear program can be solved rigorously by employing a commercial optimization package such as GINO [27]. But, these optimization packages do not offer the option of iterating among different variables as is required to come up with the global solution. To overcome these difficulties, the concept of the simplex algorithm is invoked [28–31]. The simplex algorithm can be used to generate optimal recycling solution strategies for linear programs. As part of the simplex algorithm, various artificial and slack variables are defined and the different tableaux are created. The successive transformations of the tableaux are then carried out in accordance with the various rules of the simplex method. We develop a framework of the available optimal strategies for recycling. These strategies are functions of different variables including the costs, compositions and flow rates of different sources and sinks present in the process. However, it must be noted that one of the drawbacks of the simplex algorithm is that, the number of potential solution strategies increases almost exponentially as the number of sources and sinks being considered increase. Subsequently, these strategies have been developed for three cases, which consider binary and ternary mixtures with one or two sources available for potential recycling in the process. These cases would cover a large number of potential recycling problems in industry. If necessary, similar strategies can be developed for larger number of sources and sinks based on extension of the same principles.

6. Simplex algorithm application

The determination of the cheapest possible recycling option should be taken based on consideration of both material balances and process economics. Cases 1 and 3 deal with binary systems while Case 2 considers a ternary system. Sample examples and calculations are given for Cases 1 and 2.

6.1. Case 1

This case considers two fresh species, one process source and one process sink. Let us assume that the process under consideration requires a mixed solvent consisting of two species 1 and 2. There is one process source namely, source 3 (which may have been generated via condensation, crystallization or adsorption) from solvent recovery. There are two fresh sources 1 and 2 corresponding to the fresh species 1 and 2. All compositions are given in terms of species 1.

6.1.1. Examples

1. The process sink requires that all sources being fed in add up to a total flow rate (ft) of 10 kg/s and an overall composition (xt) of 0.35.
2. The flow rate (f_3^{U}) and composition (x_3) of source 3 are 5 kg/s and 0.75 and it is obtained at a cost of C_3=US$ 0.23/kg.
3. Let fresh species 1 and 2 cost C_1=US$ 0.2/kg and C_2=US$ 0.45/kg, respectively.

The objective function is to minimize the cost of mixing, which is given by $f_1C_1+f_2C_2+f_3C_3$. Considering the cost information, it would seem that source 1 and source 3 should be used to satisfy the sink demand, as these are the cheapest sources. Another option is to recycle the recovered stream (i.e. process source 3) to its maximum and make up the rest of the sink demand by using fresh sources 1 and 2. Some of

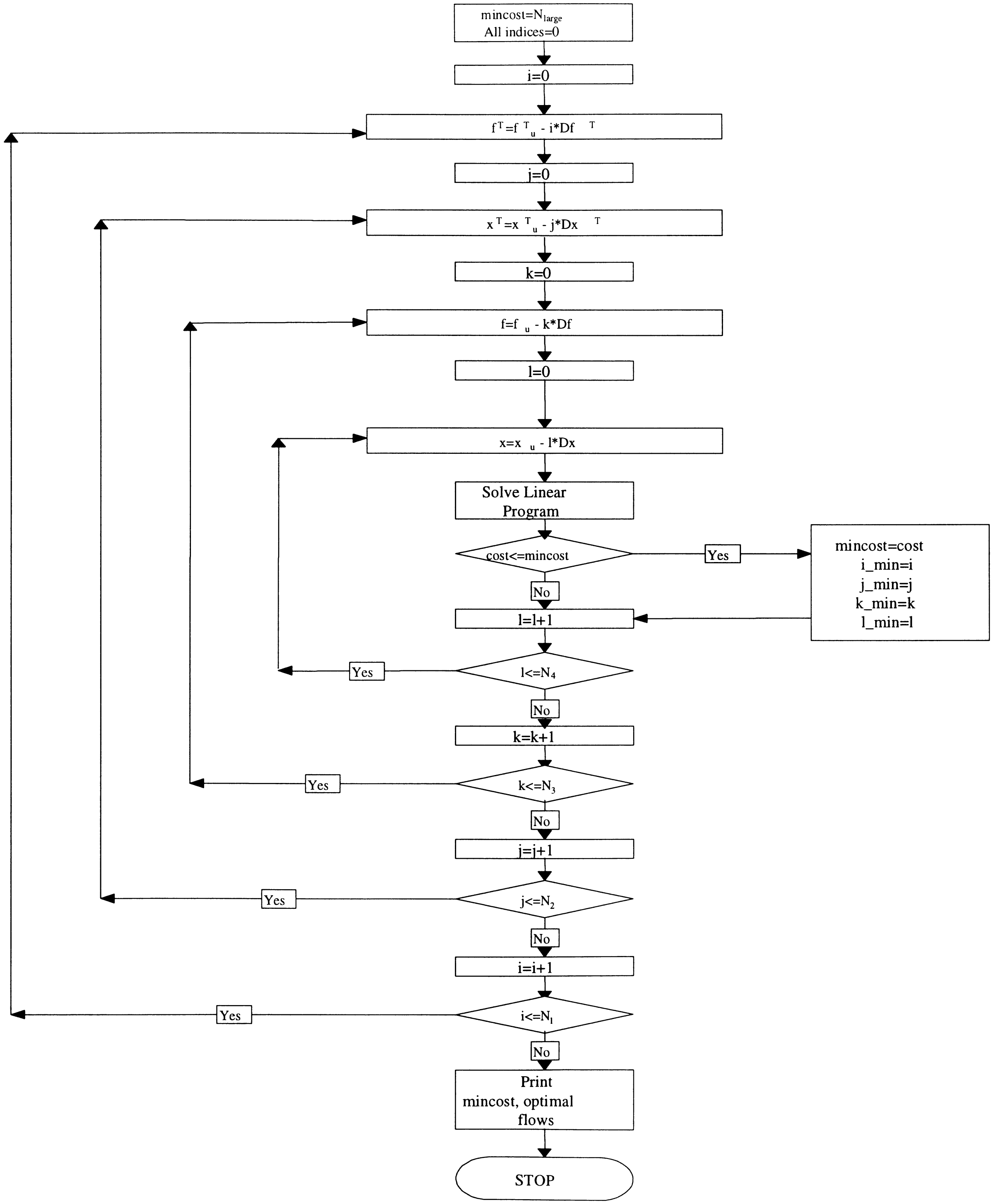

Fig. 5. Proposed solution algorithm.

the possible solutions to this problem can be summarized as follows:

1. f_3=4.0, f_1=0.5, f_2=5.5 (cost of mixing=US$ 3.495/s)
2. f_3=3.0, f_1=1.25, f_2=5.75 (cost of mixing=US$ 3.53/s)
3. f_3=1.0, f_1=2.75, f_2=6.25 (cost of mixing=US$ 3.59/s)

One can come up with more combinations to determine the minimum cost solution. However, the cheapest possible solution is to use fresh source 2 and process source 3 with flow rates f_2=5.33 and f_3=4.67 (cost of mixing=US$ 3.47/s). This solution is not obvious from either the cost data or

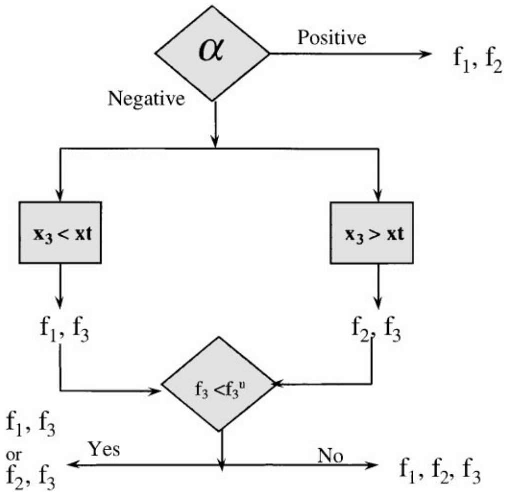

Fig. 6. Flowchart for two fresh sources and one process source.

any other information provided, but it can be obtained from Fig. 6. This flow chart has been derived using the simplex method as applied to minimization of the cost of mixing and recycling.

The relational cost parameter (α) is first calculated.

$$\alpha = (C_3 - C_2) + x_3(C_2 - C_1) \quad (29)$$

If α is positive, then the cheapest option is to use f_1 and f_2 as

$$f_1 = \text{ft} * \text{xt}, \qquad f_2 = \text{ft} * (1 - \text{xt}) \quad (30)$$

If α is negative, there are two possibilities.

Case A. $x_3 < \text{xt}$

$$f_1 = \frac{\text{ft}(\text{xt} - x_3)}{(1 - x_3)}, \qquad f_2 = 0.0, \qquad f_3 = \frac{\text{ft}(1 - \text{xt})}{(1 - x_3)} \quad (31)$$

Case B. $x_3 > \text{xt}$

$$f_1 = 0.0, \qquad f_2 = \frac{\text{ft}(x_3 - \text{xt})}{x_3}, \qquad f_3 = \frac{\text{ft} * \text{xt}}{x_3} \quad (32)$$

If the calculated f_3 value exceeds f_3^{U}, then we have to use all three sources to get the cheapest solution

$$f_1 = \text{ft} * \text{xt} - x_3 * f_3^{\text{U}},$$
$$f_2 = \text{ft} * (1 - \text{xt}) - (1 - x_3) * f_3^{\text{U}}, \qquad f_3 = f_3^{\text{U}} \quad (33)$$

Thus, in the above example, $\alpha = -0.0325$ and since $x_3 > \text{xt}$, f_2 and f_3 are used as per the above formulae. We get $f_2 = 5.33$ and $f_3 = 4.67$ and note that $f_3 \leq f_3^{\text{U}}$. Therefore, the cheapest recycling scheme can be easily determined.

6.2. Case 2

This case considers three fresh species and one process source. Let us assume that the process under consideration requires a mixed solvent consisting of three species 1, 2 and 3. There is one process source namely, source 4 (which may have been generated via condensation, crystallization or adsorption). There are three fresh sources 1, 2 and 3 corresponding to fresh species 1, 2 and 3.

6.2.1. Examples

1. The process sink requires that all sources being fed in add up to a total flow rate (ft) of 10 kg/s and an overall composition of $\text{xt}_1 = 0.35$, $\text{xt}_2 = 0.35$ and $\text{xt}_3 = 0.3$ for species 1, 2 and 3, respectively.
2. The flow rate (f_4^{U}) of process source 4 is 5 kg/s and composition of $x_1 = 0.8$, $x_2 = 0.05$ and $x_3 = 0.15$ and it is obtained at a cost of C_4=US\$ 0.15/kg.
3. Let fresh species 1, 2 and 3 cost C_1=US\$ 0.1/kg, C_2=US\$ 0.25/kg and C_3=US\$ 0.45/kg, respectively.

Considering the cost information, it would seem sources 1, 2 and 4 should be used to satisfy the sink demand, as these are the cheapest sources. Some of the possible solutions can be summarized as follows:

1. $f_1 = 2.7$, $f_2 = 3.45$, $f_3 = 2.85$, $f_4 = 1.0$ (cost of mixing= US\$ 2.565/s);
2. $f_1 = 1.9$, $f_2 = 3.4$, $f_3 = 2.7$, $f_4 = 2.0$ (cost of mixing= US\$ 2.555/s);
3. $f_1 = 1.1$, $f_2 = 3.35$, $f_3 = 2.55$, $f_4 = 3.0$ (cost of mixing= US\$ 2.545/s).

One can come up with more combinations to determine the minimum cost solution. However, the cheapest possible solution is to use fresh sources 2 and 3 with process source 4 at flow rates $f_2 = 3.28$, $f_3 = 2.34$ and $f_4 = 4.375$ (cost of mixing=US\$ 2.53/s). This solution is not obvious from the either the cost data or any other information provided, but it can be obtained from Fig. 7. This flow chart has been derived using the simplex method as applied to minimization of the cost of mixing and recycling.

The relational cost parameter (α) is first calculated as

$$\alpha = C_4 - (C_1 x_1 + C_2 x_2 + C_3 x_3) \quad (34)$$

If α is positive, then the cheapest option is to use

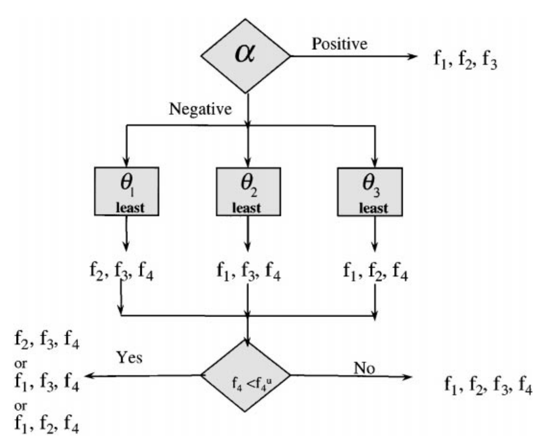

Fig. 7. Flowchart for three fresh sources and one process source.

f_1, f_2 and f_3 as

$$f_1 = \mathrm{ft} * \mathrm{xt}_1, \qquad f_2 = \mathrm{ft} * \mathrm{xt}_2,$$
$$f_3 = \mathrm{ft} * (1 - \mathrm{xt}_1) - \mathrm{ft} * \mathrm{xt}_2, \qquad f_4 = 0.0 \tag{35}$$

If α is negative then there are three possibilities. The values of θ_1, θ_2, and θ_3 are calculated as

$$\theta_1 = \mathrm{ft}\frac{\mathrm{xt}_1}{x_1}, \qquad \theta_2 = \mathrm{ft}\frac{\mathrm{xt}_2}{x_2}, \qquad \theta_3 = \mathrm{ft}\frac{\mathrm{xt}_3}{x_3} \tag{36}$$

Depending on which among these is the least value, the following solutions are obtained:

Case A. θ_1 is least

$$f_1 = 0.0, \qquad f_2 = \mathrm{ft} * \mathrm{xt}_2 - \mathrm{ft} * \mathrm{xt}_1\frac{x_2}{x_1},$$
$$f_3 = \mathrm{ft} * \mathrm{xt}_3 - \mathrm{ft} * \mathrm{xt}_1\frac{x_3}{x_1}, \qquad f_4 = \mathrm{ft}\frac{\mathrm{xt}_1}{x_1} \tag{37}$$

Case B. θ_2 is least

$$f_1 = \mathrm{ft} * \mathrm{xt}_1 - \mathrm{ft} * \mathrm{xt}_2\frac{x_1}{x_2}, \qquad f_2 = 0.0,$$
$$f_3 = \mathrm{ft} * \mathrm{xt}_3 - \mathrm{ft} * \mathrm{xt}_2\frac{x_3}{x_2}, \qquad f_4 = \mathrm{ft}\frac{\mathrm{xt}_2}{x_2} \tag{38}$$

Case C. θ_3 is least

$$f_1 = \mathrm{ft} * \mathrm{xt}_1 - \mathrm{ft} * \mathrm{xt}_3\frac{x_1}{x_3}, \qquad f_2 = \mathrm{ft} * \mathrm{xt}_2 - \mathrm{ft} * \mathrm{xt}_3\frac{x_2}{x_3},$$
$$f_3 = 0.0, \qquad f_4 = \mathrm{ft}\frac{\mathrm{xt}_3}{x_3} \tag{39}$$

If the calculated f_4 value exceeds f_4^{U}, then we have to use all four sources to get the cheapest solution.

$$f_1 = \mathrm{ft} * \mathrm{xt}_1 - x_1 * f_4^{\mathrm{U}}, \qquad f_2 = \mathrm{ft} * \mathrm{xt}_2 - x_2 * f_4^{\mathrm{U}},$$
$$f_3 = \mathrm{ft} * \mathrm{xt}_3 - x_3 * f_4^{\mathrm{U}}, \qquad f_4 = f_4^{\mathrm{U}} \tag{40}$$

Thus, in the example above, $\alpha = -0.1$ and

$$\theta_1 = 4.375, \qquad \theta_2 = 70, \qquad \theta_3 = 20$$

So, θ_1 is least and the different flow rates can be calculated as f_1=0.0, f_2=3.28, f_3=2.34 and f_4=4.375 (cost of mixing=US$ 2.53/s) and note that $f_4 \leq f_4^{\mathrm{U}}$. Therefore, the cheapest recycling scheme can be easily determined.

6.3. Case 3

This case considers two fresh species and two process sources. Suppose, a mixed solvent consisting of two species 1 and 2 is used in the process. There are two process sources, namely source 3 and 4. All compositions are given in terms of species 1. The sink requires that all sources being fed in add up to a total flow rate ft and an overall composition of xt. Let process sources 3 and 4 have flow rates of f_3^{U} and f_4^{U} while their compositions are x_3 and x_4, respectively. Let their unit costs be C_3 and C_4, respectively. There are two fresh sources 1 and 2 corresponding to the fresh species 1 and 2. Let fresh sources 1 and 2 cost C_1 and C_2, respectively. The framework for this case is given in Fig. 8. This flow chart has been derived using the simplex method as applied

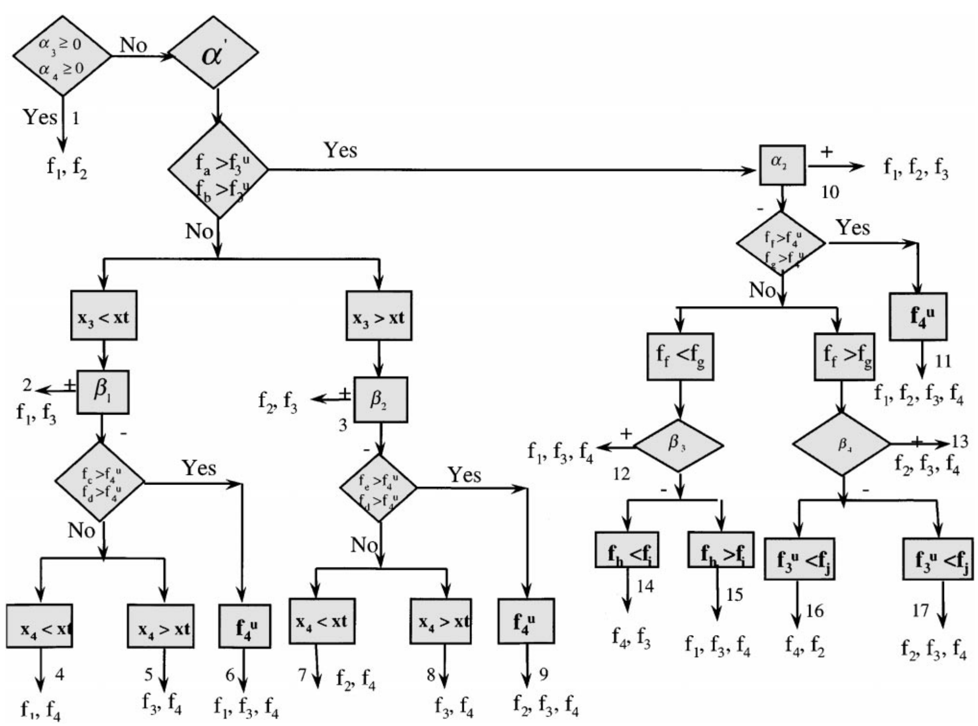

Fig. 8. Flowchart for two fresh sources and two process sources.

to minimization of the cost of mixing and recycling. The relational cost parameter (α_P) is calculated first for each process source which is given as:

$$\alpha_P = (C_P - C_2) + x_P(C_2 - C_1) \quad (41)$$

where C_p and x_p are the unit cost and composition of the respective process source. We choose source 3 as the process source with more negative of the α_Ps. So,

$$\alpha_3 = (C_3 - C_2) + x_3(C_2 - C_1),$$
$$\alpha_4 = (C_4 - C_2) + x_4(C_2 - C_1) \quad (42)$$

Also, $\alpha_3=\alpha'$. Fig. 8 can be used to get to the cheapest recycling scheme. Some of the formulae needed are listed in the Appendix A.

As can be seen from Case 3, the complexities increase rapidly as higher number of sources are considered but these three cases should be able to handle a wide variety of potential recycling schemes in typical chemical processes.

Thus, the inner level, consisting of a linear program, can be solved globally to come up with optimal recycling strategies. When coupled with the outer level iterations, this solution approach leads to global solutions to the previously defined non-linear optimization problem. We now consider the application of these principles to two case studies.

7. Case study

Case study one considers a penicillin manufacturing plant while case study two considers a urea-adduct process. Case study one utilizes the simplex rules for the binary case with one process sink as described previously. Case study two applies the simplex rules for the ternary case with one process sink as described previously. Both these case studies are examples of the application of the proposed solution algorithm to actual industrial processes.

7.1. Case study one

Given in Fig. 9 is the process flow diagram for the manufacture of penicillin [32]. The antibiotic-producing microorganism is grown in submerged culture in a fermentation medium, which contains various carbon, nitrogen and trace-metal sources required by the organism for its nutrition. The organism is grown under the conditions of pure culture, i.e. other microorganisms are excluded from the fermentation, as they might compete for nutrients. They may also produce enzymes, which are detrimental to the desired antibiotic, thus reducing the yield of the product. When the fermentation has reached peak potency, the antibiotic is recovered. Following extraction, purification and crystallization are carried out. A mixed binary solvent, consisting of methyl ethyl ketone (MEK) and methyl isobutyl ketone (MIBK) is added to the crystallization step. Following crystallization, the mother liquor is separated from the product penicillin crystals and a portion of this is recycled. Some fresh solvent is added as make-up to account for the solvent losses in the process and through the effluent. Thus, there is one process sink (crystallizer) that requires mixed solvent, one process source (from the filter) and fresh solvent, which is added as make-up.

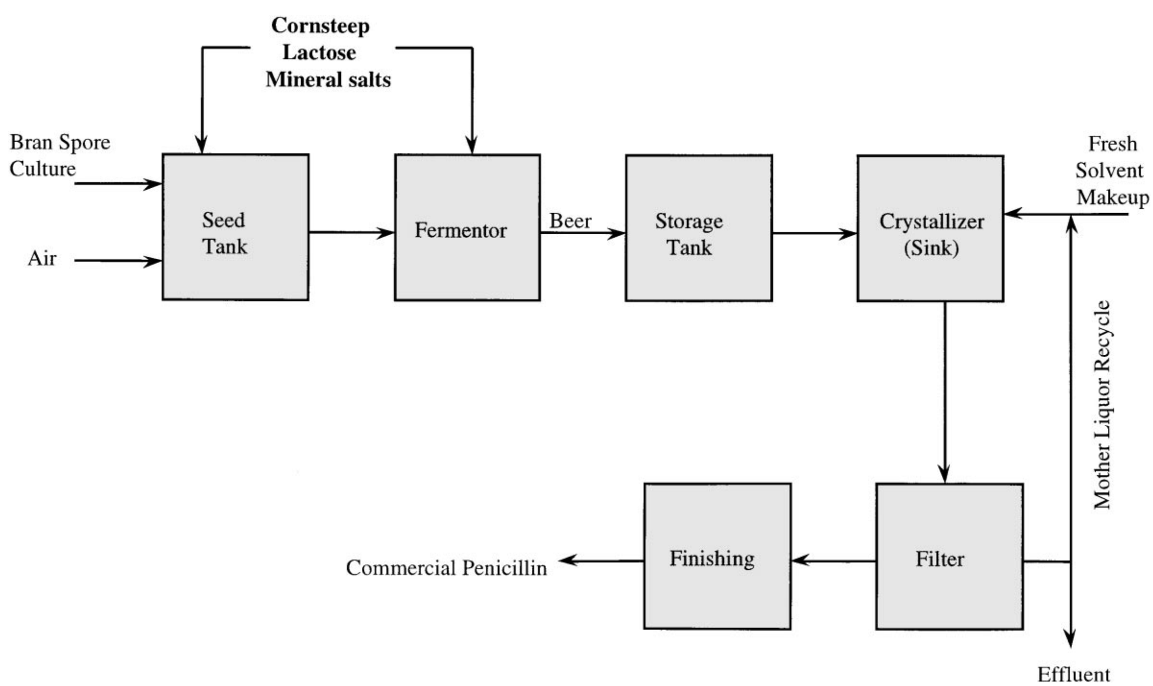

Fig. 9. Process flow diagram for the manufacture of penicillin.

The following respective upper and lower bounds have to be satisfied.

For the process sink,

$$f_u^T = 12\,\text{kg/s},\quad f_l^T = 8\,\text{kg/s},\quad x_u^T = 0.43,\quad x_l^T = 0.31$$

For the process source,

$$f_u = 7.5\,\text{kg/s},\quad f_l = 3\,\text{kg/s},\quad x_u = 0.84,\quad x_l = 0.72$$

The cost of fresh solvents MEK and MIBK are US$ 0.12 and 0.35/kg, respectively. The cost of the process source (from the filter) is US$ 0.17/kg. It is desired to find the minimum cost of mixing and recycling for the given system.

7.2. Solution to case study one

The first step in the solution is to define four increments. These are given in Eq. (25) through 28. All N values are set to 10. Four iteration indices i, j, k and l are defined for the variables f^T, x^T, f and x. The outer level in the solution algorithm consists of iterations among various values of f^T, x^T, f and x using the following:

$$f^T = f_u^T - iDf^T \tag{43}$$

$$x^T = x_u^T - jDx^T \tag{44}$$

$$f = f_u - kDf \tag{45}$$

$$x = x_u - lDx \tag{46}$$

The outer level iterations are carried out till the following conditions are satisfied:

$$i \le N_1 \tag{47}$$

$$j \le N_2 \tag{48}$$

$$k \le N_3 \tag{49}$$

$$l \le N_4 \tag{50}$$

The inner level deals with the optimization of a linear program. This case study can be solved using the framework generated earlier for a binary system with single process source and sink. The rules derived from the simplex algorithm were discussed previously (see Fig. 6). The solution algorithm is given in Fig. 5.

A FORTRAN program was written to solve the above problem. The program was run on a Sun SPARC Station 2 and converged to the optimal global solution in about 5 s. The minimum cost is determined to be US$ 1.94/s. The optimal recycle strategy would be to recycle 4.78 kg/s of the solvent recovery stream and mix with 3.22 kg/s of fresh MIBK. The final flow rate to the process sink is 8 kg/s with overall composition 0.43. The process source composition is 0.72. This solution is not apparent before the above analysis is carried out. The optimal solution recommends not using the cheapest species (MEK). Also, though up to 7.5 kg/s of a process source is available for recycle, only 4.78 kg/s is to be recycled. Thus, maximum usage of an available process source is not recommended. From the final values of the process sink and source compositions, we can calculate α using Eq. (29) as -0.0144. Using Fig. 6 and noting that x_3>xt, Eq. (32) can be used to obtain optimal flow rates of the process and fresh sources. As described previously, there exists a multitude of possible solution strategies but the above procedure ensures that the final solution is *global*.

7.3. Case study two

Given in Fig. 10 is the process flow diagram for urea-adduct separation [33]. Urea forms addition complexes with straight chain, or nearly straight chain, organic compounds such as paraffins and unsaturated hydrocarbons, acids, esters and ketones. This property allows one to separate hydrocarbons based on their affinity to form complexes with urea. The hydrocarbon feed stock is fed continuously to a stirred reactor, where it is contacted with a solution of urea in a suitable solvent. A ternary mixed solvent containing methanol, ethanol and propanol is used in this process. The reacted slurry, containing tiny crystals of urea adducts with *n*-paraffins or straight chain olefins, passes to a liquid–solid separator. The mother liquor from this goes to a settler followed by recovery of urea and solvent. After some fresh make-up is added to account for solvent losses, the recovered urea and solvent are recycled back to premixing tank and subsequently sent to the reactor. Thus, there is one process sink (premixing tank) requiring mixed solvent. There is one process source (from solvent recovery) and fresh solvent is added as make-up. This type of process is industrially highly significant and its major large-scale application is in de-waxing of lubricating oils.

The following respective upper and lower bounds are to be satisfied:

For the process sink,

$$f_u^T = 22.5\,\text{kg/s},\quad f_l^T = 19\,\text{kg/s},$$
$$x_{1,u}^T = 0.43, x_{1,l}^T = 0.3,\quad x_{2,u}^T = 0.52,\quad x_{2,l}^T = 0.37$$

For the process source,

$$f_u = 5\,\text{kg/s},\quad f_l = 1\,\text{kg/s},\quad x_{1,u} = 0.63,$$
$$x_{1,l} = 0.54,\quad x_{2,u} = 0.11,\quad x_{2,l} = 0.04$$

The cost of fresh methanol (1), ethanol (2) and propanol (3) are US$ 0.33, 0.25 and 0.37/kg, respectively. The cost of the process source (from solvent recovery) is US$ 0.29/kg. It is desired to find the minimum cost of mixing and recycling for the given system.

7.4. Solution to case study two

The first step in the solution is to define six increments corresponding to f^T, x_1^T, x_2^T, f, x_1 and x_2. These can be given

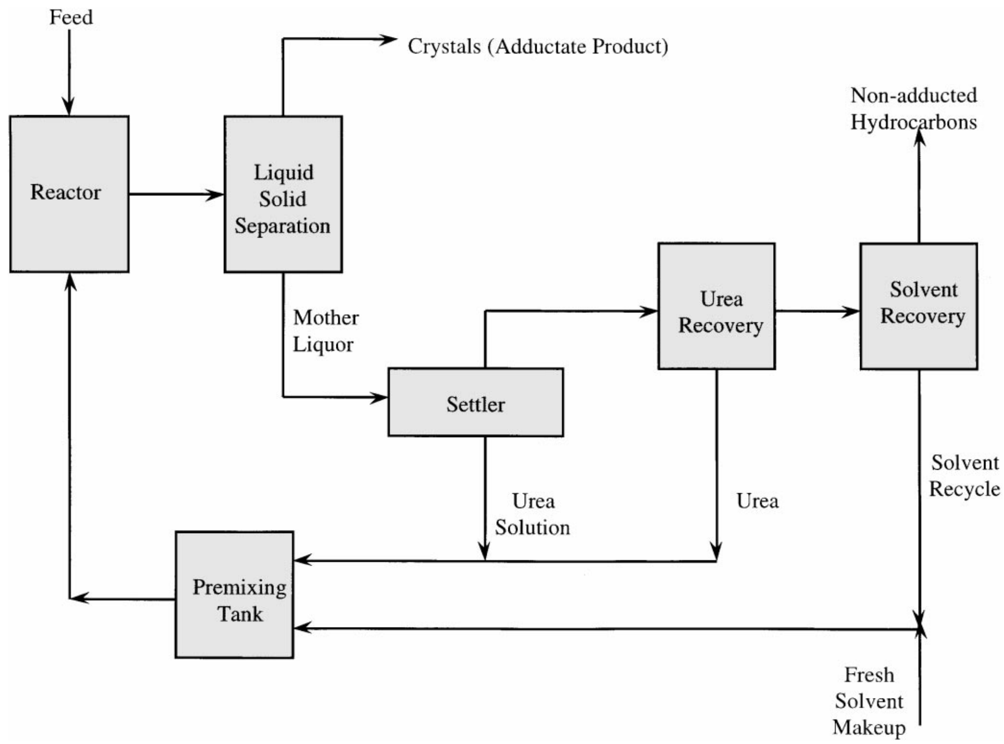

Fig. 10. Process flow diagram for the urea-adduct separation process.

as

$$\Delta f^{\mathrm{T}} = \frac{(f_{\mathrm{u}}^{\mathrm{T}} - f_{\mathrm{l}}^{\mathrm{T}})}{N_1} \quad (51)$$

$$\Delta x_1^{\mathrm{T}} = \frac{(x_{1,\mathrm{u}}^{\mathrm{T}} - x_{1,\mathrm{l}}^{\mathrm{T}})}{N_2} \quad (52)$$

$$\Delta x_2^{\mathrm{T}} = \frac{(x_{2,\mathrm{u}}^{\mathrm{T}} - x_{2,\mathrm{l}}^{\mathrm{T}})}{N_3} \quad (53)$$

$$\Delta f = \frac{(f_{\mathrm{u}} - f_{\mathrm{l}})}{N_4} \quad (54)$$

$$\Delta x_1 = \frac{(x_{1,\mathrm{u}} - x_{1,\mathrm{l}})}{N_5} \quad (55)$$

$$\Delta x_2 = \frac{(x_{2,\mathrm{u}} - x_{2,\mathrm{l}})}{N_6} \quad (56)$$

We define six iteration indices say, i, j, k, l, m and n, corresponding to the above six increments. The outer level in the solution algorithm consists of iterations among various values of $f^{\mathrm{T}}, x_1^{\mathrm{T}}, x_2^{\mathrm{T}}, f, x_1$ and x_2 using the following:

$$f^{\mathrm{T}} = f_{\mathrm{u}}^{\mathrm{T}} - i\Delta f^{\mathrm{T}} \quad (57)$$

$$x_1^{\mathrm{T}} = x_{1,\mathrm{u}}^{\mathrm{T}} - j\Delta x_1^{\mathrm{T}} \quad (58)$$

$$x_2^{\mathrm{T}} = x_{2,\mathrm{u}}^{\mathrm{T}} - k\Delta x_2^{\mathrm{T}} \quad (59)$$

$$f = f_{\mathrm{u}} - l\Delta f \quad (60)$$

$$x_1 = x_{1,\mathrm{u}} - m\Delta x_1 \quad (61)$$

$$x_2 = x_{2,\mathrm{u}} - n\Delta x_2 \quad (62)$$

Also, as all mole fractions add up to one, we have

$$x_3^{\mathrm{T}} = 1.0 - x_1^{\mathrm{T}} - x_2^{\mathrm{T}} \quad (63)$$

$$x_3 = 1.0 - x_1 - x_2 \quad (64)$$

The outer level iterations are carried out till the following conditions are satisfied:

$$i \leq N_1 \quad (65)$$

$$j \leq N_2 \quad (66)$$

$$k \leq N_3 \quad (67)$$

$$l \leq N_4 \quad (68)$$

$$m \leq N_5 \quad (69)$$

$$n \leq N_6 \quad (70)$$

The inner level deals with the optimization of a linear program. This case study can be solved using the framework generated earlier for a ternary system with single process source and sink. The rules derived from the simplex algorithm were discussed previously (see Fig. 7). A solution algorithm analogous to Fig. 5 can be set up.

A FORTRAN program was written to solve the above problem. All values of N were taken to be equal to 10. The respective increments are calculated. The program was run on a Sun SPARC Station 2 and converged to the optimal global solution in about 35 s. The optimal minimum cost was determined to be US$ 5.296/s. The optimal recycle strategy would be to recycle 4.99 kg/s of the solvent recovery stream and mix with 4.33 kg/s of fresh methanol and 9.68 kg/s of fresh ethanol. The final flow rate to the process sink is 19 kg/s with compositions of methanol, ethanol and propanol being 0.39, 0.52 and 0.09, respectively. The process source compositions were 0.62, 0.04 and 0.34 for methanol, ethanol and propanol, respectively. This solution is not apparent before the above analysis is carried out.

From the final values of the process sink and source compositions, we can calculate α (from Eq. (34)) as -0.0504. Using Fig. 7, the values of θ_1, θ_2 and θ_3 are calculated as 11.95, 247 and 4.99, respectively (from Eq. (36)). As θ_3 has the least value, Eq. (39) will give the optimal flow rates of the process and fresh sources. It must be noted that the above solution approach, besides coming up with the optimal flow rates of various sources to be recycled, also fixes the optimal compositions of the process sources and sinks. This enables the identification of an optimal operating point for the process at which the cost of mixing and recycling would be minimized.

8. Conclusions

This paper considers an important aspect of every chemical process. It proposes a solution strategy to come up with the *global* cheapest mixing and recycling scheme. A generic non-linear program formulation is given and the solution algorithm, consisting of two different levels, is proposed. The decomposition into two levels was made based on the observation that the non-linear program reduces to a linear program in a special case. The solution strategy is generic enough to handle many sources and sinks. The simplex method is used to generate solutions for cases with lower number of sources and sinks. It is used to derive frameworks wherein, the optimal flow rates of the different sources can be obtained as functions of relative costs and compositions. These frameworks can be used to generate optimal recycling schemes. A relational cost parameter is introduced and used. Two different case studies, dealing with the manufacture of penicillin and a urea-adduct process, are considered. The *global* recycling solution was determined for each case study and it was shown that these solution strategies are not arbitrary but are generated based on the systematic application of the proposed solution algorithm.

Acknowledgements

The author would like to thank Dr. Mahmoud El-Halwagi for his insight into this research work.

Appendix A

Some of the formulae used in Fig. 8 are listed below

$$\beta_1 = \alpha_2 - \alpha_1 \frac{(1-x_4)}{(1-x_3)}$$

$$\beta_2 = \alpha_2 - \alpha_1 \frac{x_4}{x_3}$$

$$\beta_3 = \alpha_2 \frac{(1-x_3)}{(1-x_4)} - \alpha_1$$

$$\beta_4 = \alpha_2 \frac{x_3}{x_4} - \alpha_1$$

$$f_\mathrm{a} = \mathrm{ft}\frac{(1-\mathrm{xt})}{(1-x_3)}$$

$$f_\mathrm{b} = \mathrm{ft}\frac{\mathrm{xt}}{x_3}$$

$$f_\mathrm{c} = \mathrm{ft}\frac{(1-\mathrm{xt})}{(1-x_4)}$$

$$f_\mathrm{d} = \mathrm{ft}\frac{(\mathrm{xt}-x_3)}{(x_4-x_3)}$$

$$f_\mathrm{e} = \mathrm{ft}\frac{\mathrm{xt}}{x_4}$$

$$f_\mathrm{f} = f_\mathrm{c} - f_3^\mathrm{U}\frac{(1-x_3)}{(1-x_4)}$$

$$f_\mathrm{g} = f_\mathrm{e} - f_3^\mathrm{U}\frac{x_3}{x_4}$$

$$f_\mathrm{h} = f_3^\mathrm{U} - \mathrm{ft}\frac{(x_4-\mathrm{xt})}{(x_4-x_3)}$$

$$f_\mathrm{i} = f_3^\mathrm{U} - f_\mathrm{a} + f_4^\mathrm{U}\frac{(1-x_4)}{(1-x_3)}$$

$$f_\mathrm{j} = f_3^\mathrm{U} - f_\mathrm{b} + f_4^\mathrm{U}\frac{x_4}{x_3}$$

From Fig. 8, there are 17 possible outcomes depending on various conditions listed above and each of these is marked with a number. The flow rates can be calculated as

1. $f_1 = \mathrm{ft}(1-\mathrm{xt}), \qquad f_2 = \mathrm{ft} * \mathrm{xt}$
2. $f_1 = \mathrm{ft}\frac{(\mathrm{xt}-x_3)}{(1-x_3)}, \qquad f_3 = \mathrm{ft}\frac{(1-\mathrm{xt})}{(1-x_3)}$
3. $f_2 = \mathrm{ft}\frac{(x_3-\mathrm{xt})}{x_3}, \qquad f_3 = \mathrm{ft}\frac{\mathrm{xt}}{x_3}$
4. $f_1 = \mathrm{ft}\frac{(\mathrm{xt}-x_4)}{(1-x_4)}, \qquad f_4 = \mathrm{ft}\frac{(1-\mathrm{xt})}{(1-x_4)}$
5. $f_3 = \mathrm{ft}\frac{(x_4-\mathrm{xt})}{(x_4-x_3)}, \qquad f_4 = \mathrm{ft}\frac{(\mathrm{xt}-x_3)}{(x_4-x_3)}$

6. $f_1 = \mathrm{ft}\dfrac{(\mathrm{xt}-x_3)}{(1-x_3)} - f_4^{\mathrm{U}}\dfrac{(x_4-x_3)}{(1-x_3)}$,

$f_3 = \mathrm{ft}\dfrac{(1-\mathrm{xt})}{(1-x_3)} - f_4^{\mathrm{U}}\dfrac{(1-x_4)}{(1-x_3)}, \qquad f_4 = f_4^{\mathrm{U}}$

7. $f_2 = \mathrm{ft}\dfrac{(x_4-\mathrm{xt})}{x_4}, \qquad f_4 = \mathrm{ft}\dfrac{\mathrm{xt}}{x_4}$

8. $f_3 = \mathrm{ft}\dfrac{(x_4-\mathrm{xt})}{(x_4-x_3)}, \qquad f_4 = \mathrm{ft}\dfrac{(\mathrm{xt}-x_3)}{(x_4-x_3)}$

9. $f_2 = \mathrm{ft}\dfrac{(x_3-\mathrm{xt})}{x_3} + f_4^{\mathrm{U}}\dfrac{(x_4-x_3)}{x_3}$,

$f_3 = \mathrm{ft}\dfrac{\mathrm{xt}}{x_3} - f_4^{\mathrm{U}}\dfrac{x_4}{x_3}, \qquad f_4 = f_4^{\mathrm{U}}$

10. $f_1 = \mathrm{ft} * \mathrm{xt} - x_3 * f_3^{\mathrm{U}}$,

$f_2 = \mathrm{ft}(1-\mathrm{xt}) - (1-x_3)f_3^{\mathrm{U}}, \qquad f_3 = f_3^{U}$

11. $f_1 = \mathrm{ft} * \mathrm{xt} - x_3 * f_3^{\mathrm{U}} - x_4 * f_4^{\mathrm{U}}$,

$f_2 = \mathrm{ft}(1-\mathrm{xt}) - (1-x_3)f_3^{\mathrm{U}} - (1-x_4)f_4^{\mathrm{U}}$,

$f_3 = f_3^{\mathrm{U}}, \qquad f_4 = f_4^{\mathrm{U}}$

12. $f_1 = \mathrm{ft}\dfrac{(\mathrm{xt}-x_4)}{(1-x_4)} + f_3^{\mathrm{U}}\dfrac{(x_4-x_3)}{(1-x_4)}$,

$f_3 = f_3^{\mathrm{U}}, \qquad f_4 = \mathrm{ft}\dfrac{(1-\mathrm{xt})}{(1-x_4)} - f_3^{\mathrm{U}}\dfrac{(1-x_3)}{(1-x_4)}$

13. $f_2 = \mathrm{ft}\dfrac{(x_4-\mathrm{xt})}{x_4} + f_3^{\mathrm{U}}\dfrac{(x_3-x_4)}{x_4}$,

$f_3 = f_3^{\mathrm{U}}, \qquad f_4 = \mathrm{ft}\dfrac{\mathrm{xt}}{x_4} - f_3^{\mathrm{U}}\dfrac{x_3}{x_4}$

14. $f_3 = \mathrm{ft}\dfrac{(x_4-\mathrm{xt})}{(x_4-x_3)}, \qquad f_4 = \mathrm{ft}\dfrac{(\mathrm{xt}-x_3)}{(x_4-x_3)}$

15. $f_1 = \mathrm{ft}\dfrac{(\mathrm{xt}-x_3)}{(1-x_3)} - f_4^{\mathrm{U}}\dfrac{(x_4-x_3)}{(1-x_3)}$,

$f_3 = \mathrm{ft}\dfrac{(1-\mathrm{xt})}{(1-x_3)} - f_4^{\mathrm{U}}\dfrac{(1-x_4)}{(1-x_3)}, \qquad f_4 = f_4^{\mathrm{U}}$

16. $f_2 = \mathrm{ft}\dfrac{(x_4-\mathrm{xt})}{x_4}, \qquad f_4 = \mathrm{ft}\dfrac{\mathrm{xt}}{x_4}$

17. $f_2 = \mathrm{ft}\dfrac{(x_3-\mathrm{xt})}{x_3} + f_4^{\mathrm{U}}\dfrac{(x_4-x_3)}{x_3}$,

$f_3 = \mathrm{ft}\dfrac{\mathrm{xt}}{x_3} - f_4^{\mathrm{U}}\dfrac{x_4}{x_3}, \qquad f_4 = f_4^{\mathrm{U}}$

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