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Genetic Algorithm Optimization of Fractional Crystallization Processes

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Abstract

This paper advances the optimization of fractional crystallization separation flowcharts applying genetic algorithms (GA), using as example the potassium nitrate separation. All feasible separation sequences are described using the thermodynamic state network model The optimization criterion is the minimization of the sum of flows over the entire network. The results obtained are compared with the solution found by solving the non-linear optimization problem implemented in GAMS.

Keywords Optimization**,** genetic algorithms, salt separation, fractional crystallization

1. Introduction

In most of the industrial applications referring to salt separation by fractional crystallization, sequences of heating, vaporization, cooling, and solid phase separation units are used. The design of complex fractional crystallization systems and the subsequent selection of a good solution is quite complicated due to the multitude of separation pathways. Most of the traditional research in this field relies on the identification and generation of separation schemes on the phase diagrams [1, 7]. More recently, Cisternas and Swaney [2] developed a flow sheet network based methodology to optimize the sequencing design of fractional crystallization process. In this method, the liquid-solid equilibrium furnishes data for an accurate evaluation of the relative solubility variation at

different possible working conditions representing multiple saturation points. The feasible pathways in the separation process are represented in a network. The network is generated according to the general rules of separation sequences synthesis applied to fractional crystallization [1]. According to these rules, the n-components system is divided in all (n-1)-components systems, which are again divided in all (n-2)-components systems and so on until two solutes systems are reached. A network is derived for each system based on thermodynamic considerations. The overall network is an aggregate of all networks considered as feasible solutions. The flows and unknown compositions are then calculated by formulating an optimization problem. Some other further developments of the network concept applied for fractional crystallization optimization have been extended for more complex cases where a task network is also defined and heat integration is included [4,5].
The objective of the present work is to evaluate the capability of heuristic search methods (GA) in finding the optimal design of a crystallization based separation scheme. To solve this problem we used the thermodynamic state network particularly applied for KNO_3 crystallization.

2. Problem formulation

To formulate the mathematical model for the separation system, the topology of the state network is basically described by the definition of the nodes and arches as presented in [2, 3] and already used in previous work [6]. The equations of the mathematical model are component mass balances in operation nodes (multiple saturation points and intermediate solute product nodes). Additional restriction referring to the feed, product and intermediate solute products are also considered.

2.1. Thermodynamic state network definition for KNO_3 crystallization

Potassium nitrate can be obtained by the double decomposition reaction:

$$KCl + NaNO_3 \Leftrightarrow KNO_3 + NaCl$$

The products in this system are separated by fractional crystallization. This process was previously analysed using the equilibrium diagram [7, 1] and using the state network methodology and optimization in the frame of GAMS software [6]. In this system, KNO_3 is the valuable product, while NaCl is a waste. $NaNO_3$ is also a valuable raw material, and is recycled to the reaction step. Alike previous work [6], we consider that KCl is totally consumed by the chemical reaction and we seek to separate by fractional crystallization a solution of 19.8% KNO_3, 11.5% NaCl, 3.3% $NaNO_3$ and 65.4% water, which is an eutectic solution taken as reference point in feeding. The equilibrium data for the quaternary reciprocal system were taken from [8], cited in [1] and presented in table 1. Only two working temperatures were chosen for the product separations: 20°C and 100°C. At these temperatures the relative solubility of the

salts in the working points varies at enough large extent that all separations are possible. From technical point of view both values of the temperatures can be easily realized.

Table 1. Solubility data for KNO_3-NaCl-$NaNO_3$-H_2O system

Node	Temp, °C	Saturated solution, wt%			Solid phase
		KNO_3	NaCl	$NaNO_3$	
C1	20	15.68	11.20	27.02	KNO_3+NaCl+$NaNO_3$
H1	100	49.96	2.88	32.74	KNO_3+NaCl+$NaNO_3$
C2	20	21.31	21.56	0.0	KNO_3+NaCl
H2	100	61.14	10.92	0.0	KNO_3+NaCl
C3	20	0.0	13.80	30.34	NaCl+$NaNO_3$
H3	100	0.0	5.63	57.40	NaCl+$NaNO_3$
C4	20	17.13	0.0	42.03	KNO_3+$NaNO_3$
H4	100	48.10	0.0	36.70	KNO_3+$NaNO_3$

Points H1 and C1 correspond to the quaternary eutectic points of the KNO_3-NaCl-water system. Points H2-H4 and C2-C4 correspond to ternary eutectics in three of the four ternary systems associated to quaternary reciprocal system. The generation of feasible structures relies on the evaluation of the relative compositions in a two solute system according to the procedure described in [1, 2]. The solving of a three solute system is similar, successively considering each salt a pseudo-solvent component and thus identifying the possible separation points of intermediate solute products. The deriving of all feasible pathways for this system is presented in details in [6]. The final thermodynamic state network is characterized by 17 nodes and 54 arches and is presented in figure 1. Water can be added or withdrawn from any multiple saturation node.

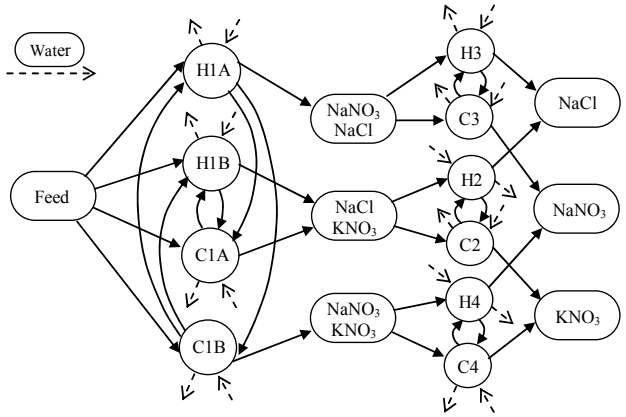

Figure 1. Thermodynamic state network for the three solute separation system

2.2. The mathematical model

As co-precipitation is allowed, and the composition of intermediate products is unknown, the component mass balances in all 17 nodes forms a nonlinear system of equations. The unknowns are the flowrates L_i, i =1…54 and the intermediate product compositions. The total number of unknowns is thus 60. The system consist of 47 equations (16 for component mass balances in the three-solute multiple saturation nodes, 18 for the component mass balances in the two-solutes saturation nodes, 4 equations of component mass balance defining the feed split in hot and cold points, 3 mass balance equations for intermediate products, 3 mass balance equations for final products, 3 equations defining the normalization of intermediate product composition). The objective function attached to this model is defined as the sum of flows in the separation scheme.

3. Optimization procedure

The independent variables was selected in order to transform the system of equations defined by mass balances in a linear one, solved by MATLAB function *lsqr*. Consequently, 3 values of intermediate product concentrations bounded in the domain [0 1], and 10 flowrates values bounded between 0 and 100 (the feed flowrate was taken 100 units) define the set of independent variables. These correspond to the chromosomes of the genetic algorithm.
The genetic algorithm used is a MATLAB implementation [9] that can be downloaded at ftp://ftp.eos.ncsu.edu/pub/simul/GAOT. Float representation of chromosomes has been used. The selection of candidate chromosomes for crossover and mutation is made according with a ranking selection function based on the normalized geometric distribution. Three types of crossover are applied: simple, interpolated, and extrapolated crossover. In the simple crossover, the crossover point is randomly selected. The interpolated crossover performs an interpolation along the line formed by the two parents. The extrapolated crossover performs an extrapolation along the line formed by the two parents in the direction of better parent. Four types of mutation are applied: boundary, multi-nonuniform, nonuniform, and uniform mutation. Boundary mutation changes one gene of the selected chromosome randomly either to its upper or lower bound. Multi-nonuniform mutation changes all genes, whereas nonuniform mutation changes one of the genes in a chromosome on the base of a non-uniform probability distribution. This Gaussian distribution starts wide, and narrows to a point distribution as the current generation approaches to the maximum generation. Uniform mutation changes one of the genes based on a uniform probability distribution. The numbers of applications of the different crossover and mutation operators are imposed as parameters of the genetic algorithm. Their default values have been used, respectively for each generation: 2 simple, 2 interpolated, and 2 extrapolated crossover, 4 boundary, 6

multi-nonuniform, 4 nonuniform, and 4 uniform mutation. Due to the use of a maximization algorithm, the chromosome fitness corresponds to the negative value of the objective function. This was a penalty function, respectively to the real objective (the sum of the flow rates) was added the sum of the weighted absolute values of the residuals of equations multiplied with a penalty coefficient having the value 10^8. Some values of the calculated flowrates (with MATLAB function *lsqr*) have negative values. If these values correspond to water draw flows in a node, the negative value was assigned to an opposite sense of water in/out operation and corrected by assigning the absolute value to the dilution water entering the node. Similar considerations were made for dilution water flows. For negative recycle flows the chromosome was abandoned by assigning a big value to the objective function.

4. Results and discussions

The same optimum solution as that given by GAMS [6] was obtained after about 200 generation The size of initial population was 1000 chromosomes, and the maximum number of generations was set to 1000. The total computing time was 2 s on a computer with Intel Pentium 4, 1.73 GHz processor and 512 MB DDR 533 memory. This solution (figure 2) is reported and described in details from technological point of view in [6].

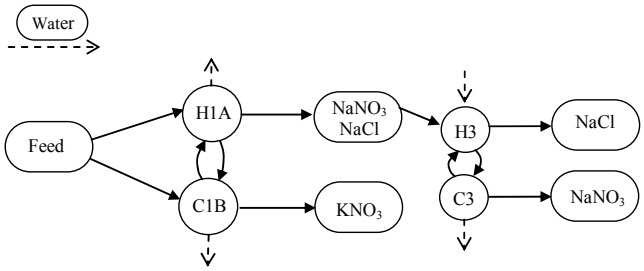

Figure 2. Network representation obtained by optimisation procedure in GAMS and by GA

During several other trials an other optimal solution was generated corresponding to a lower value of the objective function (figure 3).
Although a smaller value of the objective function was reached, the analysis of the corresponding separation flowchart proves that it is not necessarily a better solution from technological point of view. The difference representing the total flow reduction (about 5 %) is not significant to justify a supplementary heating unit. This result will be considered in future analyses of this separation system, and other objective functions including the number and nature of required operating units will be formulated.

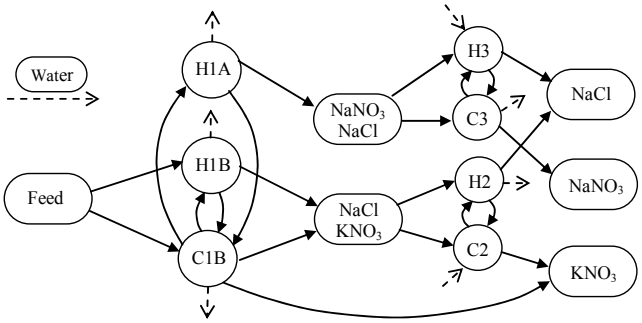

Figure 3. Optimal network corresponding to minimum flows

In both solutions obtained in this work the feed is divided between the hot and cold operating point, unlike similar solution obtained in [1]. This feed split allows smaller internal flows without salting out operations.

5. Conclusions and future work

The results of this study prove the good ability of genetic algorithms to find optimal solutions, in a reasonable computing time, for the separation sequence optimization. As compared with NLP problem solved in GAMS, the final results are similar. GA could automatically avoid the gap of infeasible solution due to large population and possible large number of generations. In GAMS implementation, feasible solutions were reached by running the application with different starting points and convenient variable boundaries. Concerning the problem formulation, further work is required. More restrictions referring to the number and type of operating units should be defined, and some other possible objective functions could be derived.

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