Design of Optimal Process Flowsheet for Fractional Crystallization Separation Process

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ABSTRACT: A procedure is presented that synthesizes fractional crystallization separation processes to obtain pure solids from multi-component solutions. The method includes a procedure to generate a network flow model to identify alternative process designs for fractional crystallization. The main advantage of this systematic procedure with respect to other reported procedures is using non-equilibrium solubility values for crystallizers and mixing tanks as process points. By employing an approach factor to show the distance from equilibrium, a non-linear programming model is obtained. Solving the non-linear programming model, optimal process flowsheet and the corresponding non-equilibrium solubility values for process points are presented. The proposed procedure is used to design an optimal flowsheet for production of sodium carbonate and sodium sulfate from Burkeite. The results show that when the approach factor goes to unity, the maximum efficiency of separation process is attained which corresponds to the minimum total flow rates and total cost of the process.

KEY WORDS: Fractional crystallization, Process synthesis, flowsheet, Non-linear programming model.

INTRODUCTION

Crystallization is an attractive alternative for separating close boilers, high melting compounds, and heat sensitive materials that are difficult or impossible to separate by distillation. Crystallization is applied extensively in the chemical industry, both as a purification process and a separation process. It is extensively used in different industrial applications, including the production of a wide range of materials such as fertilizers, detergents, foods, and pharmaceutical products, as well as in the treatment of waste effluents. However, compared to the extensive works on multi-

component distillation process design, relatively little works are available on multi-component crystallization. This is serious omission because wide applications of multi-component crystallization in the industry. This problem causes that most other chemical processes can be simulated and optimized on computer today, but the same is not true for processes involving crystallization especially for multi-component electrolyte mixtures. It is important to note that the worldwide annual production rate of fertilizer chemicals such as ammonium phosphates and ammonium nitrate each exceed one million-ton.

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Some of the above mentioned salts are produced by fractional crystallization, a process in which several salts are separated as pure phases from a multi-component mixture.

Fractional crystallization effects separations by manipulating the relative solubilities of the components in a solution using operations such as heating or cooling, evaporative crystallization, dissolving, stream combining, reaction crystallization and salting-out by injection of supercritical fluid. The technique is used to separate a wide variety of chemicals. For example, minerals such as borax, lithium chloride monohydrate, potash, sodium carbonate, and sodium chloride are crystallized from brines. Despite the significance of fractional crystallization of salts, little information is available on the conceptual design of these processes. Fractional crystallization is expected to play a more significant role in future separations because of the greater need to minimize waste disposal and to reduce the loss of desirable products.

the limited publications on crystallization design, Fitch (1970) [1] explained a systematized approach for conceptual design of fractional crystallization processes for systems in which the crystalline species do not form solid solutions. He used graphical method to represent the system, its equilibria, and the results of various process manipulations. He proposed synthesis methods for fractional crystallization processes using various types of solid-liquid equilibrium based phase diagram and assumed solubility to be a linear function of temperature. Cisternas and Rudd (1993) [2] developed material balances for the separation techniques discussed in Fitch (1970) [1]. This is important because the costs for the plant are directly related to the flow rates in the process. A limitation of all the designs is that calculation of phase diagrams and solubilities is not addressed and the effects of complex-ionic equilibrium on design equations are not considered.

Liu et al. (2007) [3] studied the fractional crystallization of carnallite in pure water, KCl aqueous solution and MgCl₂ aqueous solution with experimental observations. They indicated that when carnallite (KMgCl₃.6H₂O) was dissolved in pure water, porous crystals of KCl were formed. However, when the fractional crystallization occurred in KCl or MgCl₂ aqueous solution, the solid crystals of KCl with different morphologies were formed. They concluded that the

coupling of a high dissolution rate of carnallite and a high nucleation rate of KCl nearby the surface of carnallite resulted in the porous crystal of KCl.

Rajagopal, Ng and Douglas (1988, 1992) [4,5] developed Douglas's hierarchical procedure for solid processes based on heuristics and short-cut models. Their proposed procedure is limited to low-molecular-weight solids. Since the short-cut models are used in their program, their results are only good for obtaining quick, preliminary estimates of the best alternatives.

Rajagopal, Ng and Douglas (1991) [6] gave a procedure for the separation of a binary mixture by means of extractive crystallization so that it can be applied for a given chemical system. Dye and Ng (1995a) [7] discussed the use of extractive crystallization to completely separate two and three solutes from a solution, respectively. A similar approach was taken for the conceptual design of fractional crystallization processes (Dye and Ng, 1995b) [8]. Furthermore, Ng (1991) [9] provided a systematic method to separate a multi-component mixture of molecular solids.

Lin et al. (2007) [10] presented the synthesis and design of crystallization-based separation processes for systems involving solid solution in the presence of a solvent. They used solid-liquid equilibrium phase diagrams in designing the flowsheet and determination of operating conditions.

Berry and Ng (1997) [11] recognized the need for phase diagrams at various temperatures for design of fractional crystallization processes, but their method required a prior knowledge of activity coefficients. Berry and Ng (1996) [12] gave a design method to synthesize fractional crystallization processes for separating conjugate salts from systems composed of simple and compound salts.

Takano et al. (2002) [13] presented a methodology for conceptual design and analysis of crystallization-based separation processes involving electrolyte systems. They classified the activity coefficient models for electrolyte solutions. They also explained the application of the thermodynamic part and the integration of thermodynamic part with the flowsheet design/analysis part. But they did not imply the detail of their methodology to integrate the thermodynamic part and design/analysis part.

Wibowo and Ng (2002) [14] extended Douglas' hierarchical procedure for crystallization and solids

processing as an illustration of workflow management for process synthesis and development.

Thomsen et al. (1995, 1998) [15, 16] outlined a general method for the calculation of various types of phase diagrams for aqueous electrolyte mixtures integrated with simulation, design and optimization of fractional crystallization. They applied the extended UNIQUAC model for describing the aqueous electrolyte systems. They did not describe of their methodology to combine phase diagrams for aqueous electrolyte mixtures with process simulation, design and optimization of fractional crystallization processes.

Cisternas et al. (1999, 1998 and 2004) [17-19] presented a network flow model for synthesizing crystallization-based separations for multicomponent systems. They assumed that the composition of components between unit operations (e.g. crystallizers and mixing tanks) were in the thermodynamic equilibrium states. With this assumption a linear programming model is obtained They determined the desired process flowsheet that incorporated the effects of cake washing operations [19].

Above described publications on fractional crystallization show that there are two major approaches for the synthesis of crystallization-based separation. In one approach, the phase equilibrium diagram is used for the identification of separation schemes (For example Cisternas and Rudd, 1993; Lin et al., 2007 and Berry et al., 1997) [2, 10,11]. While these procedures are easy to understand, they are relatively simple to implement only for simple cases. For more complex systems, such as multi-component systems and multiple temperatures of operation, the procedure is difficult to implement because the graphical representation is complex and because there are many alternatives to study. The second strategy is based on simultaneous optimization using mathematical programming based on a network flow model between feasible thermodynamic states [17-19].

In this work the network flow model is used to identify of alternative process designs for fractional crystallization. Since non-equilibrium solubility values are used for crystallizers and mixing tanks, a non-linear programming model is obtained. By solving the non-linear programming model, optimal process flowsheet and the corresponding non-equilibrium solubility values for unit operations are presented.

MODEL DEVELOPMENT

The network flow model proposed by Cisternas et al. [17, 18] is used to identify of alternative process designs for fractional crystallization. They assumed that the crystallizers and dissolving tanks as process points were worked in equilibrium states which this assumption led to a linear program model. The main advantage of our systematic procedure is using non-equilibrium solubility values for crystallizers and mixing tanks as process points. Since non-equilibrium solubility values for process points are added to design variables, our optimization problem is a non-linear programming model. An approach factor (η) is applied to show the distance from equilibrium. Solving the non-linear programming model, optimal process flowsheet and the corresponding non-equilibrium solubility values for process points are presented. The optimization program seeks process flowsheet with low rates of recycle, evaporation and dilution. The model is constructed based on the following assumptions:

- 1- The model is applied to systems with two salts and one solvent, including multiple feeds, multicomponent products, and systems that form one or more multiple salts.
- 2- Operations in the network flow model may include dissolution, crystallization, solids separation, solvent removal and mixing.
- 3- The general form of process is represented in an overall network of nodes and arcs. Nodes are corresponding to a practical process points, solute intermediate species, process feeds, and end products. Solute intermediate species arise when the phase equilibrium gives solid phases that are not end products (e.g., double salts). Arc represents the material flows between nodes.
- 4- Since many industrial crystallization processes do not attain equilibrium, in this work for the first time, multiple saturation points are not used as process points. For instance, in *Perry et al.*, a 95 % approach to equilibrium condition is reported as normally possible with inorganic salts [20]. Therefore, non-equilibrium solubility values should be used in place of equilibrium diagrams.
- 5- The compositions of the crystallizers and of the tanks used for dissolution or dilution as well as all the flow rates are design variables.

6- Investment and operating costs of fractional crystallization systems are increasing functions of the evaporating, dilution, and recycle flow rates.

Set of nodes is defined as:

 $S=\{s | all nodes in the system\}$

This set includes practical process points, solute intermediate species, process feeds, and end products. The components including solutes, and solvents are defined by the set $I=\{i\}$. The arcs, which denotes streams between nodes, are denoted by $L=\{1\}$. The following subsets are:

 $L_N=\{l|l\in L, \text{ solvent, feeds, solute intermediate streams with known compositions }\}$

$$\begin{split} L_{\text{UN}} = & \{l|l \in L, \text{streams with composition not specified}\} \\ S_F = & \{s|s \in S, \text{ feed nodes with flow rates of specie } i \in I \text{ specified}\} \end{split}$$

 $S_P \!\!=\!\! \{ \ s | s \in S \text{, product nodes with flow rates of specie} \\ i \ \in I \ \text{specified} \ \}$

 $S_M \!\! = \!\! \{s | s \in S, \text{ intermediate solute product of specie } i \in I \text{ specified } \}$

 $S_S=\{ s|s \in S, \text{ process points with unknown composition} \}$

 $S^{in}(s) = \{1 | 1 \in L, \text{ is an inlet stream to node } s, s \in S\}$

 $S^{out}(s) = \{l | l \in L, \text{ is an outlet stream to node } s, s \in S\}$

$$S_{N}^{in}$$
 (s)= S^{in} (s) $\cap L_{N}$

 S_N^{out} (s)= S^{out} (s) $\cap L_N$

 S_{IJN}^{in} (s)= S^{in} (s) $\cap L_{IJN}$

 S_{UN}^{out} (s)= S^{out} (s) $\cap L_{UN}$

 $F(s) = \{ | l | l \in L, \text{ is an outlet from node } s, s \in S_F \}$

 $P(s) = \{ l | l \in L, \text{ is an outlet from node } s, s \in S_P \}$

 $I_P(s) = \{i | i \in I, \text{ is a component specified in node } s, s \in S_P \}$

 $I_F(s) = \{ \ i | i \in I, \text{ , is a component specified in node } s, \, s \\ \in \, S_F \, \}$

Each stream 1 is associated with the variable mass flow rate w_l and with the composition of each component. $x_{l,i}$ is composition of component i of stream 1, if the composition is known such as the compositions of solvent, feeds, solute intermediate streams and products. $z_{l,i}^{noneq}$ is composition of component i of stream 1 in nonequilibrium state, if the composition is unknown.

Using the above sets, parameters, and variables that describe the overall network for the separation problem,

the constraints are defined as follows:

Mass balances for process point nodes in steady state condition:

$$\sum_{l \in S^{\text{in}}(s)} w_l - \sum_{l \in S^{\text{out}}(s)} w_l = 0 \qquad s \in S_M$$
 (1)

Mass balances for each component in process point nodes:

$$\sum_{l \in F(sl)} \hspace{-0.2cm} w_l \cdot x_{l,h} + \hspace{-0.2cm} \sum_{l \in S_{UN}^{in}(s2)} \hspace{-0.2cm} w_l \cdot z_{l,i}^{noneq} - \hspace{-0.2cm} (2)$$

$$\sum_{l \in P(s3)} \!\!\! w_l \cdot x_{l,j} - \!\!\!\! \sum_{l \in S_M(s4)} \!\!\!\! w_l \cdot x_{l,k} - \!\!\!\! \sum_{l \in S_{UN}^{out}} \!\!\!\! w_l \cdot z_{l,i}^{noneq} = \!\!\! 0$$

where $s1 \in S_F$, $h \in S_F$, $s2 \in S_s$, $i \in I$, $s3 \in S_p$, $j \in I_p(s3)$, $s4 \in S_M$, $k \in I_M(s4)$

Specification for feeds flow rates:

$$\sum_{l \in F(s)} w_l.x_{l,i} = C_{s,i}^F \qquad s \in S_M, i \in I_F(s)$$
 (3)

where $C_{s,i}^F$ is the desired flow rates of specie i in feed s.

Specification for product flow rates:

$$\sum_{l \in P(s)} w_{l}.x_{l,i} = C_{s,i}^{P} \quad s \in S_{P}, i \in I_{P}(s)$$
(4)

where $C_{s,i}^{P}$ is the desired quantity of component i in final product s.

Equality of composition for the inlet and outlet streams of the intermediate product nodes:

$$x_{1,i} = x_{m,i} \ l \in S_M^{in}(s), m \in S_M^{OUT}(S), s \in S_M$$
 (5)

Non-negativity and composition constraints:

$$1 \ge z_{1,i}^{\text{noneq}} \ge 0 \quad ; \quad w_1 \ge 0 \tag{6}$$

Since many industrial crystallization processes do not attain equilibrium, therefore, dynamic phase diagrams (non-equilibrium solubility values) should be used in place of equilibrium diagrams. The composition of component i of stream 1 from the operating nodes in non-equilibrium state, z_{li}^{noneq} , is related to the composition of component i of stream 1 in equilibrium state, x_{li}^{eq} , by following set of equations:

Nodes	Composition of nodes		odes	
	na2co3	na2so4	h2o	Solid phase
3	0.1495	0.112	0.7385	$Na_2CO_3.10H_2O + Na_2SO_4.10H_2O$
4	0.1495	0.112	0.7385	$Na_2CO_3.10H_2O + Na_2SO_4.10H_2O$
5	0.1790	0.162	0.659	$Na_2CO_3.10H_2O + Na_2SO_4.10H_2O$
6	0.1790	0.162	0.659	$Na_2CO_3.10H_2O + Na_2SO_4.10H_2O$
7	0.2580	0.086	0.656	$Na_{2}CO_{3}.10H_{2}O + Na_{2}CO_{3}.2Na_{2}SO_{4}$
8	0.2580	0.086	0.656	$Na_{2}CO_{3}.10H_{2}O + Na_{2}CO_{3}.2Na_{2}SO_{4}$
9	0.1550	0.195	0.650	Na ₂ CO ₃ .2Na ₂ SO ₄ + Na ₂ SO ₄
10	0.1550	0.195	0.650	Na ₂ CO ₃ .2Na ₂ SO ₄ + Na ₂ SO ₄
11	0.2970	0.055	0.648	$Na_2CO_3.H_2O + Na_2CO_3.2Na_2SO_4$
12	0.2970	0.055	0.648	$Na_2CO_3.H_2O + Na_2CO_3.2Na_2SO_4$
13	0.1140	0.222	0.664	Na ₂ CO ₃ .2Na ₂ SO ₄ + Na ₂ SO ₄
14	0.1140	0.222	0.664	Na ₂ CO ₃ .2Na ₂ SO ₄ + Na ₂ SO ₄

Table 1: Solubility data [21].

$$\frac{z_{l,i}^{noneq} - x_{l,i}^{eq}}{x_{l,i}^{mixture} - x_{l,i}^{eq}} 1 - \eta \tag{7}$$

where η is an approach factor that the state of crystallizers or mixing tanks to be closer to equilibrium state. In the literature, a 95 % approach to equilibrium condition is reported as normally possible with inorganic salts [20], but the approach factor, η , is lower than 0.95 in industrial applications. $x_{1,i}^{mixture}$ is the composition of component i in the mixture before any crystallization or dissolution occurs. $x_{1,i}^{mixture}$ is obtained by:

$$x_{l,i}^{nixture} = \frac{\sum_{l \in F(s2)} w_{l} \cdot x_{l,i} + \sum_{l \in S_{un}^{in}(s1)} w_{l} \cdot z_{i,j}^{noneq} + \sum_{l \in S_{M}(s4)} w_{l} \cdot x_{l,k}}{\sum_{l \in F(s2)} w_{l} + \sum_{l \in S_{un}^{in}(s1)} w_{l} + \sum_{l \in S_{M}(s4)} w_{l}}$$
(8)

The objective function minimizes the cost. The following equation can be used as an objective function.

$$\min_{\mathbf{w}_{1},\mathbf{z}_{1,i}} \sum_{\mathbf{s} \in \mathbf{S}} \mathbf{C}_{\mathbf{s}} \cdot \left[\sum_{\mathbf{l} \in \mathbf{S}^{\text{in}}(\mathbf{s})} \mathbf{w}_{1} \right]^{\alpha} + \tag{9}$$

$$\sum_{s \in S_F} C_s \cdot \left\lfloor \sum_{l \in F(s)} w_l \right\rfloor + \sum_{s \in S_P} C_s \cdot \left\lfloor \sum_{l \in P(s)} w_l \right\rfloor$$

where C_s is the weighted cost coefficient. The first term on the left-hand side of Eq. (9) represents the annual capital cost, and the second and third terms represent the annual operation cost of raw material and products,

respectively. Product costs count for solvent evaporation and residue loss. For the purposes of this study, all the C_s are equal to 1 and α =1. With this simplicity, the optimization program seeks process flowsheet with low rates of feed, product, recycle, evaporation and dilution.

Thus, the objective function in Eq. (9) subject to constraints in Eqs. (1) to (8), defines a nonlinear programming problem in which the variables to be optimized are mass flow rates w_l and the unknown compositions $z_{l,i}$.

RESULTS AND DISCUSSION

In this paper, a procedure is discussed that synthesizes fractional crystallization separation processes to obtain pure solids from multi-component solutions. The fractional crystallization design problem can be stated as a network flow model which represents all the possible set of potential separation flow sheet structures [17-19]. The main advantage of our systematic procedure is using non-equilibrium solubility values for crystallizers and mixing tanks. Since non-equilibrium solubility values for process points are added to design variables, our optimization problem is a non-linear programming model.

Let us consider a system as an example to reveal our proposed procedure. This example considers the production of sodium carbonate and sodium sulfate from 100 t per day of Burkeite (Na₂CO₃.2Na₂SO₄). Data for the phase diagram of Na⁺-CO₃²⁻-SO₄²⁻-H₂O are available in table 1 [21]. Fig. 1 shows the flow network for the present example. This flowsheet is constructed as follows: Nodes 1 and 2 are included for feed and water,

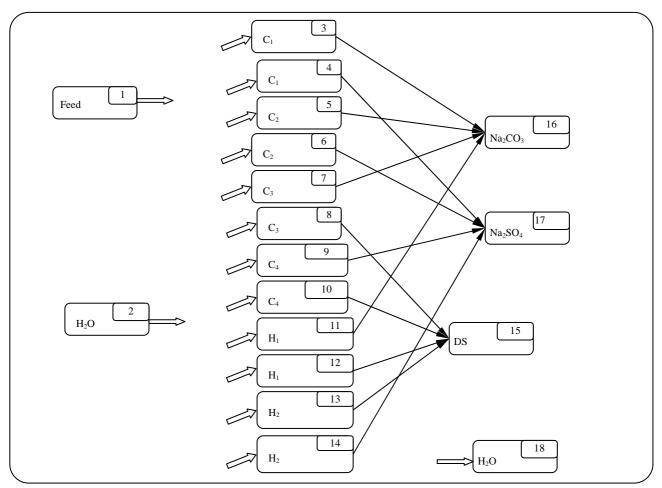


Fig. 1: The flow network for the Burkeite separation process.

which can be added to nodes 3 to 14. Nodes 3 to 14 are included for the potential process points (crystallizers and dissolving tanks). If a potential operating point could be used to produce more than one salt, a node must be included for each salt. Node 15 shows the double salt Burkeite. Nodes 16 and 17 are for sodium carbonate and sodium sulfate products, respectively.

For this procedure, the problem corresponds to a nonlinear programming problem, because the compositions of the process points (nodes 3 to 14) are unknown. The compositions of the other nodes are known. The variables in the synthesis are the flow rates between nodes and the compositions of each process points. This problem has 89 independent equations and 116 parameters. The interconnection among the nodes (arcs) could not be drawn in the diagram. Fig. 1 includes only the arcs that represent salt production. The open arrows represent the feasible interconnection paths that

have been omitted for clarity. This program minimizes the Eq. (9) by taking into account the process constraints (Eqs. (1) to (8)). Fig. 2 demonstrates the flow structure obtained by solving non-linear programming problem for η values equal 1, 0.95, 0.9 and 0.7. Fig. 3 gives the conceptual design of the process for η values equal 1, 0.95 and 0.9. The obtained flow rates correspond to n for four values of 1, 0.95, 0.9 and 0.7 are shown in tables 2 to 5. In these tables, w_{i,j} represents the flow rate from node i to node j. Fig. 4 shows the stream mass flow rates for different flows versus approach factor (n). F is total flow rates in the flowsheet (Eq. (9)). As shown in Fig. 5, it can be noted that total flow in the flowsheet, F, increases as the approach factor η decreases. Therefore when the approach factor, η , goes to 1, the maximum efficiency of separation process is attained which corresponds to the minimum total flow rates and minimum total cost of the process.

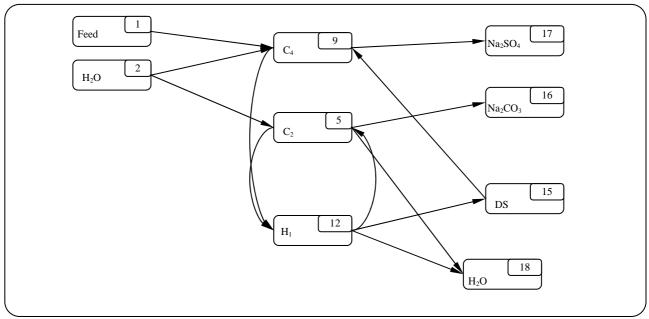


Fig. 2: The flow structure design for the Burkeite separation process for η values equal 1, 0.9, 0.95 and 0.7.

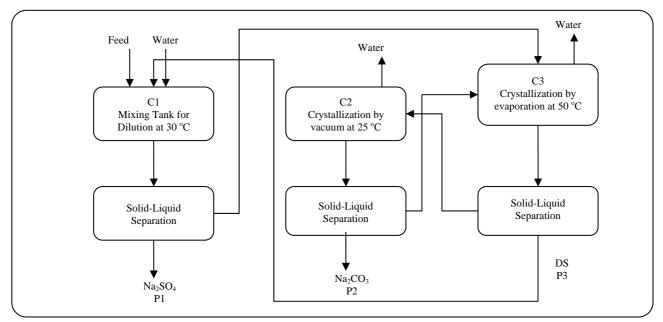


Fig. 3: The conceptual design of the process for η values equal 1, 0.95 and 0.9.

In Fig. 3, the feed stream and the recycle stream of double salt (DS) are fed to mixing tank. This tank is operated at 30 $^{\circ}$ C as a dilution tank and salt Na₂SO₄ is removed as the product. The mother liquor is then mixed with the recycle stream from vacuum crystallizer at 25 $^{\circ}$ C. This mixture is fed to an evaporative crystallizer. This crystallizer operates at 50 $^{\circ}$ C and separates double salt (Na₂CO₃.2Na₂SO₄).

The mother liquor from evaporative crystallizer forms the feed to the vacuum crystallizer. This crystallizer works at 25 $^{\circ}$ C with salt Na₂CO₃.10H₂O being the solid product. Note that this flowsheet is obtained by optimization program which seeks process flowsheet with low rates of recycle, evaporation and dilution.

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Table 2: Crystallizer Temperatures, Node Compositions and Stream Mass Flow Rates for $\eta = 1$. F=1290.7.

Crystallizer	Operating Temperature
C1	30 °C
C2	25 °C
C3	50 °C

Nodes	Composition of nodes		
	na2co3	na2so4	h2o
1	0.2717	0.7283	0
2	0	0	1
3	0.1495	0.112	0.7385
4	0.1495	0.112	0.7385
5	0.179	0.162	0.659
6	0.179	0.162	0.659
7	0.258	0.086	0.656
8	0.258	0.086	0.656
9	0.155	0.195	0.65
10	0.155	0.195	0.65
11	0.297	0.055	0.648
12	0.297	0.055	0.648
13	0.114	0.222	0.664
14	0.114	0.222	0.664
15	0.2717	0.7283	0
16	0.3704	0	0.6296
17	0	0.4409	0.6296
18	0	1	0
19	0	0	1

Stream	Mass flow
$W_{1,9}$	100
$W_{2,5}$	0
W _{2,9}	214.718
W _{9,12}	330.33
W _{5,12}	39.05
W _{12,5}	115.018
W _{9,17}	72.83
W _{5,16}	73.35
W _{5,18}	2.6151
W _{12,15}	88.447
W _{15,9}	88.447
W _{12,18}	165.92

Table 3: Crystallizer Temperatures, Node Compositions and Stream Mass Flow Rates for η =0.95. F=1363.5.

Crystallizer	Operating Temperature
C1	30°C
C2	25 °C
C3	50 °C

	Composition of nodes		
Nodes	na2co3	na2so4	h2o
1	0.2717	0.7283	0
2	0	0	1
3	0.1549	0.1092	0.7385
4	0.1424	0.1067	0.7385
5	0.1846	0.157	0.659
6	0.1702	0.1541	0.659
7	0.2585	0.0847	0.656
8	0.2528	0.0918	0.656
9	0.1536	0.2023	0.65
10	0.1532	0.1986	0.65
11	0.29	0.0621	0.648
12	0.1133	0.2243	0.664
13	0.2717	0.7283	0
14	0.3704	0	0.6296
15	0	0.4409	0.6296
16	0	1	0
17	0	0	1
18	0.2717	0.7283	0
19	0	0	1

Stream	Mass flow
W _{1,9}	100
W _{2,5}	0
W _{2,9}	223.9773
W _{9,12}	347.72
W _{5,12}	49.4985
W _{12,5}	125.1815
W _{9,17}	72.83
W _{5,16}	73.35
W _{5,18}	2.3296
W _{12,15}	96.5733
W _{15,9}	96.5734
W _{12,18}	175.4647

Table 4: Crystallizer Temperatures, Node Compositions and Stream Mass Flow Rates for η =0.9. F=1447.

Crystallizer	Operating Temperature
C1	30 °C
C2	25 °C
C3	50 °C

	Composition of nodes		
Nodes	na2co3	na2so4	h2o
1	0.2717	0.7283	0
2	0	0	1
3	0.1582	0.1073	0.7385
4	0.1362	0.1022	0.7385
5	0.1894	0.1528	0.659
6	0.1613	0.1461	0.659
7	0.257	0.0838	0.656
8	0.2478	0.0978	0.656
9	0.1522	0.2095	0.65
10	0.1537	0.2094	0.65
11	0.2831	0.0696	0.648
12	0.1127	0.2268	0.664
13	0.2717	0.7283	0
14	0.3704	0	0.6296
15	0	0.4409	0.6296
16	0	1	0
17	0	0	1
18	0.2717	0.7283	0
19	0	0	1

Stream	Mass flow
W _{1,9}	100
W _{2,5}	0
W _{2,9}	234.2657
W _{9,12}	367.04
W _{5,12}	62.942
W _{12,5}	138.1032
W _{9,17}	72.83
W _{5,16}	73.35
W _{5,18}	1.8077
W _{12,15}	105.6029
W _{15,9}	105.6029
W _{12,18}	186.275

Table 5: Crystallizer Temperatures, Node Compositions and Stream Mass Flow Rates for η =0.7. F=1982.

Crystallizer	Operating Temperature
C1	30 °C
C2	25 °C
C3	50 °C

Nodes	Composition of nodes		
	na2co3	na2so4	h2o
1	0.2717	0.7283	0
2	0	0	1
3	0.1648	0.103	0.7385
4	0.1433	0.1074	0.7385
5	0.2008	0.1436	0.659
6	0.1258	0.1143	0.659
7	0.1825	0.061	0.656
8	0.2247	0.1316	0.656
9	0.1466	0.2386	0.65
10	0.1538	0.2477	0.65
11	0.2562	0.1025	0.648
12	0.1101	0.2366	0.664
13	0.2717	0.7283	0
14	0.3704	0	0.6296
15	0	0.4409	0.6296
16	0	1	0
17	0	0	1
18	0.2717	0.7283	0
19	0	0	1

Stream	Mass flow
$W_{1,9}$	100
W _{2,5}	4.4767
W _{2,9}	290.1165
W _{9,12}	471.907
W _{5,12}	171.73
W _{12,5}	240.6067
W _{9,17}	72.83
W _{5,16}	73.35
W _{5,18}	0
W _{12,15}	154.6203
W _{15,9}	248.41
W _{12,18}	4.4767

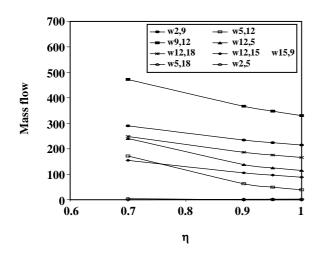


Fig. 4: Stream mass flow rates for different flows versus approach factor (η).



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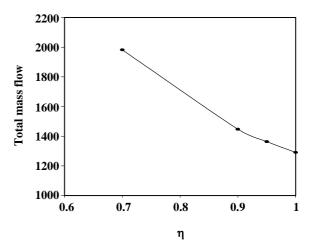


Fig. 5: Total mass flow versus approach factor (η) .

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