# TESCE, Vol.29, No.2 pp. 175 -1944 FUZZY APPROACH FOR SEPARATION SEQUENCE SYNTHESIS

#### S.ALY

Suez Canal University, Faculty of Petroleum and Mining Engineering, Department of chemicaJ and Refining Engineering. Suez-Egypt

#### **Abstract**

A new approach based on fuzzy set theory for the synthesis of sharp separations is presented. The synthesis algorithm is based on the application of expert rules and consists of the three steps: qualification of the estimate separation mass load coefficients, difference in normal boiling points, relative volatility, and separation ratio between distillate and bottom in a fuzzy rule based procedure- the classical conjunction of fuzzy compatibility degrees-and the choice of the best separation point. This approach has been tested for a number of conventional problems reported in literature. The results of the proposed method are consistent with the reported optimum solution and need much less computation in comparison with some algorithms published.

*Keywords:* Distillation, fuzzy set, Heuristic

#### **1. Introduction**

Distillation based separation sequences exist in almost all-chemical processes. Such separation systems are used for feed preparation, product separation and finishing as well as for waste treatment. Due to the significant contribution of the distillation sequences to the capital and operating cost of the total chemical process, the development of a systematic framework which will select the optimum distillation sequence becomes an important research issue.

Many published works have dealt with the synthesis of sharp separation sequences in the last two decades. The main papers in this area are reviewed in Hendry et al. [1], Hlavacek [2], Nishida et al. [3], and Westerberg [4]. Following these authors, the methods proposed for solving sharp separator sequence synthesis can be classified into three main categories: heuristic

approaches, evolutionary strategies, and algorithmic methods. The heuristic approach, which uses rule of thumb that are based on engineering judgment .or experience, was developed first. Most of the proposed heuristics were classified into groups including: composition heuristics, separation factor heuristics, and separation technique heuristics [5]. Although the reduction of the search space of solutions by the heuristic approach may be quite important, an optimal solution is not guaranteed. Furthermore, many of the known heuristics contradict or overlap each other [6]. Evolutionary strategies, which try to identify the best process through a sequence of evolutionary improvements, include the three following subtasks: generate an initial process by means of heuristics, define the evolutionary rules, and determine the evolutionary strategy. The evolutionary approach depends essentially upon both the initial flowsheet generated and the evolutionary strategies that can be classified into two main categories. The heuristic strategy [7], where the rules are selectively chosen and the algorithmic strategy [8], where breadth first or depth first techniques are used. The disadvantage of the evolutionary method, which is strategy dependent, is that it needs a good initial sequence. Algorithmic methods based upon mathematical programming techniques may consist of thousands of linear and nonlinear equations containing both discrete and continuous variables [9]. Mathematical programming based design seeks to develop and optimize a superstructure to the design space. This necessarily restricts design consideration to the proposed superstructure. Therefore, the development of an automated design system, based on this approach, would require the specification of all such as dynamic programming [10], branch and bound [11], and mixed integer linear programming [12], show several essential disadvantages.

- By means of dynamic programming no straightforward stream recycles can be treated.
- The efficiency of branch and bound based approaches depends mainly on the discovery of reasonable upper bounds for the annual costs
- The formulation of a mathematical program to solve a practical problem may require a substantial investment of time.

In this work, a fuzzy approach based on the fuzzification of four heuristic rules will be used for the selection of optimum separation sequence .The proposed procedure combines the values of the estimate separation mass load coefficients, the difference in normal boiling points, relative volatility, and separation ratio between distillate and bottom for components in a fuzzy rule base procedure. The proposed method will be tested for a number of synthesis problems, which have been solved previously using other approaches,

#### **2- Problem description**

The problem to be addressed can be stated as follows:

*Given a single midticomponent feed mixture of known conditions (Le. flowrate, composition, temperature and pressure) synthesize a process that can isolate the desired products from the feed with a minimal annual cost*" The main assumptions for this synthesis problem are the following:

- Only simple (single feed ,two product streams) straight distillation columns are considered without energy integration;
- Each column operates at high recovery (>98%), sloppy splits of key components (non sharp separation) are not allowed ;
- Mixture or division of intermediate stream are prohibited;
- Saturated liquid feeds are present in each distillation column;
- Component volatility order dose not changes in the sequence.

Most of the investigators in the field of separation sequence synthesis have chosen this set of assumptions, indeed this choice allows a direct comparison of our result with other published works.

#### **3-Fuzzy** set **theory**

Zadeh first formulated fuzzy set theory in 1965 [13], The theoretical information's are available in Dubois and Prade [14]. We will just explain the basic notions of such theory and the typical applications in chemical engineering. Fuzzy set theory is able to describe uncertainty that can arise in a lot of manner in chemical engineering. Following Kraslawski [15], we distinguish two main kinds of uncertainty: ambiguity and imprecision. A proposition is ambiguity if its truth, or its falsity, can not be definitely established. A proposition is imprecise if its value is not sufficiently determined with respect to a given scale. Both ambiguity and imprecision can be also divided into many uncertainty types [15]. Here, the uncertainty of our heuristic rules are on the latter type, because of the lack of precision of terms like " high" or " small".

. A fuzzy set A in the space  $X=\{x\}$  can be defined as the set:

$$
A = \{x, \mu_A(x)\} \text{........ } \forall x \in X
$$

$$
\mu_A: X \to [0,1] \text{........ } \text{........ } (1)
$$

$$
x \to \mu_A(x)
$$

 $\mu_A(x)$  Expresses the grade of membership of *x* in *A*.  $\mu_A(x)=0$  Means that *x* is definitely a member of *A*;  $\mu_4(x)=1$  Means that x is definitely a member of A.

The intermediate values of the membership function denote partial defined to some extent, membership of *A.* 

The fuzzy set theory is, in effect, a step toward a rapprochement between the precision of classical mathematics and the pervasive imprecision of the real world, a rapprochement born of the incessant human quest for a better understanding of mental processes and cognition (see e.g. Zadeh, [16]). Some algebraic operations can be defined on fuzzy set [14] like:

Union 
$$
\mu_{A \cup B} = \max (\mu_A, \mu_B)
$$
........ (2)

Intersection *<sup>M</sup> = mm(/JA , jUB* ) (3 )

A decision is to be made by evaluating all the related rules at different levels in a knowledge base. The evaluations are carried out according to the MAX-MIN algorithm (see e.g. Zimmermann, [17]),

$$
\mu_j(x) = \max_{i \in I} \left\{ \min_{k \in K} \{ \mu_{i1}(x_1), \mu_{i2}(x_2), ..., \mu_{ik}(x_k), ..., \mu_{iK}(x_k) \} \right\} \dots \dots (4)
$$

Where  $\mu_k(x)$  = membership function of variable x in fuzzy set *k* representing the *k*th antecedent of the *i*th rule at the *j*th level,

 $\mu_i(x)$  = Membership function of variable x in the fuzzy set pertaining to the rule selected to be fired at the jth level and  $x, x_k$ = variables.

The **MAX-MIN** algorithm is implemented in two stages. The antecedent of a rule can be represented by the truth value expression:

$$
\tau_i = \min\{\mu_{i1}(x_1), \mu_{i2}(x_2), ..., \mu_{ik}(x_k), ..., \mu_{ik}(x_k)\}........(5)
$$

The **MIN** operation yields a set truth values  $(\tau_i)$  through evaluation of the membership functions of all the rules. Then, a single rule is selected by performing the MAX operation, i.e.

 $\tau = \max \{\tau_1, \tau_2, ..., \tau_j\}.$  (6)

This selected rule is activated or fired. The same operation is repeated at the succeeding level based on the information received from the preceding level.

#### **4-Quantification of the rules**

The synthesis algorithm is based on the application of expert rules, well suited for an economical design problem. From an extensive compilation of Aly *et al.* [18], four rules of thumb have been retained:

1) Favour the separation where the difference  $\Delta T_h$  of boiling point temperatures between two adjacent key components is the most important

2) Favour the separation at the point where the relative volatility  $\alpha_{i,j}$  of two adjacent key components is the most important.

3) Favour equimolar separation between the distillate D and the bottom B. The ratio D/B or B/D must be as close as possible to one.

4) Favour the separation where the estimated mass load coefficient (EML) is less important. This coefficient EML is the molar flowrate that has to be processed by all separation units in the downstream sequence. It is a linear function of molar fractions of each component in the mixture (see Appendix 1)

From a physical point of view, it can be observed that rules 1 and 2 partially overlap, because they both privilege the separation point where the physical properties exploited in the separation method are maximum, and consequently the cost minimum. Rules 3 and 4 are both related to the mass load to be separated. However, they are not totally redundant, insofar as the EML coefficient takes into account the total mass load to be separated from the current separation step up to the complete separation, when rule 3 is only related to the current separation step. These four rules incorporate the two well known precepts in the synthesis of separation trains [19] : "The most delicate separations must be carried out last" and " separations that eliminate the most abundant components are preferable".

These four rules are expressed in vague and imprecise terms like " important" or" close to one .As mentioned by Dubois and prade [14] , the fuzzy set theory offers a general framework for representing uncertainty and vagueness. The above rules are quantified by fuzzy quantities represented by the following membership functions:

Rule 2 
$$
\mu_2 = \begin{cases} 0 & \text{if } \alpha_{i,j} \prec 1.1 \\ \frac{\alpha_{i,j} - 1.1}{0.9} & \text{if } 1.1 \le \alpha_{i,j} \le 2 \text{...,} \end{cases}
$$
 (8)

Rule 3 
$$
\mu_3 = \begin{cases} P \dots & \text{if } 0 \le P \le 1 \\ (2 - P) \dots & \text{if } 1 \le P \le 2 \dots & \dots \dots (9) \\ 0 \dots & \text{otherwise} \end{cases}
$$

Where P corresponds to the ratio D/B or B/D

Rule 4  
\n
$$
\mu_4 = \begin{cases}\n0 & \text{...} \\
EM_{\text{max}} - EML \\
EML & \text{...} \\
EM_{\text{min}} \le EML \le EML \\
1 & \text{...} \\
\end{cases}
$$
\n
$$
if EML + EML
$$
\n
$$
EML + EML
$$
\n
$$
EML + EML
$$

TESCE, Vol.29, No.2

#### 5-Fuzzy approach strategy

The algorithm which is followed in this paper is to select the best separation sequence which consists of three sequential as shows in fig. LThe strategy is based on the evaluation, for each possible split, of the validity of each rule. After the step of quantification, the values of  $\mu_1$ ,  $\mu_2$ ,  $\mu_3$  and  $\mu_4$  for each split i are Calculated and then, we apply:

 $\mu_{rule1}$  *AND* $\mu_{rule2}$  *AND* $\mu_{rule3}$  *AND* $\mu_{rule4}$  = min( $\mu_{1}$ ,  $\mu_{2}$ ,  $\mu_{3}$ ,  $\mu_{4}$  ) (decreases) (1) and

$$
\mu^* = \max_i (\mu_{split,i}) = \max_i (\min(\mu_1, \mu_2, \mu_3, \mu_4)_{split,i}). \dots \dots \dots (12)
$$

and we choose the split corresponding to The main steps of the strategy are summarized in Fig.1.



Fig .1 Main steps of the fuzzy strategy.

# **6. Illustrative example**

Let us consider the separation of a 5-component mixture attributed to Wankat [20]. The problem specification is given in Table 1.

radio 11. Se component separation system				
Component	Mole	Normal boiling	Relative	
	fraction	point difference	volatility	
A: Ethanol	0.25			
		4.3	1.093	
B: i-Propanol	0.15			
		15.3	1.862	
C: n-Propanol	0.35			
		11.2	1.349	
D: i-Butanol	0.10			
		8.5	1.449	
E: n- Butanol	0.15			

Table 1: 5-component separation system

(1) The EML and P ( ratio D/B or B/D) are calculated for each split



(2) The values of grades of membership are evaluated for each possible split.



TESCE , Vol.29, No.2 -183- -183- July 2003

 $(3)$ We apply

$$
\mu^* = \max_i \left( \mu_{split,i} \right) = \max_i \left( \min_i \left( \mu_1, \mu_2, \mu_3, \mu_4 \right)_{split,i} \right)
$$
  

$$
\mu^* = \max_i \left( \min_i \left( 0.000, 0.333, 0.000, 0.330 \right) \right)
$$
  

$$
\mu^* = \max_i \left( \min_i \left( 1.000, 0.667, 0.847, 0.650 \right) \right)
$$
  

$$
\min_i \left( 1.000, 0.333, 0.277, 0.550 \right)
$$
  

$$
\min_i \left( 0.744, 0.177, 0.388, 0.243 \right)
$$
  

$$
\mu^* = \max_i \left( 0.000, 0.650, 0.277, 0.177 \right) = 0.650
$$

That corresponds to the split AB/CDE (4) The procedure is repeated with the mixture CDE

 $EML=0.417$  $EML=0.750$ P=0.715 P=0.333 for CD/E for C/DE  $\mu_{\text{I}} = 1.000$   $\mu_{\text{2}} = 0.715$   $\mu_{\text{3}} = 0.277$   $\mu_{\text{4}} = 0.583$  for C/DE  $\mu_{\text{F}}$  0.000  $\mu_{\text{2}}$  = 0.333  $\mu_{\text{3}}$  = 0.388  $\mu_{\text{4}}$ =0.250 for CD/E **/**   $\mu$  = max  $\min($ 1.000,0.715,0.277,0.583)  $\,$  \ , min(0.000,0.333,0.388,0.250) **V:**   $\mu^*$  = max (0.277, 0.000) = 0.277

We then choose the split C/DE the resulting sequence **[AB/CDE,C/DE,A/B,D/EJ** is the optimal one (see Table 2) these results are consistent with optimum solution for the same problem reported by Wankat [20 ] and Flowers et al.[21 ]

TESCE, Vol.29, No.2 184- 184- July 2003



### Table 2: Possible separation sequence for 5-component Separation system [21]

# **7-Comparison with other methods**

### *Example 1: Separation of a six-component mixture*

Consider the separation of a mixture of a mixture of light olefins and paraffin's by ordinary distillation. The mixture as originally presented by Thompson and King [19] is given in Table 3.

Table 3: Example 1 data

Component	Mole fraction	Normal boiling	Relative
		point difference (k)	volatility
A: Ethane	0.20		
		40.9	3.50
B: Propylene	0.15		
		5.7	1.20
C: Propane	0.20		
		35.8	2.70
D: I-Butane	0.15		
		5.8	1.12
E: n-Butane	0.15		
		36.5	3.00
$F: n$ -pentane	0.15		

A summary of the results of the proposed method is shown in Table 4 . Thus the resulting sequence is **[ABC/DEF,A/BC,DE/F, B/C, D/E** ] . A comparison, on the same problem, of our work with these of Nadgir and Liu [22], Nath and Motard [7] and Seader and Westerberg [8] shows that our result corresponds to the best sequence found by the heuristic method of Nadgir and Liu and by the heuristic evolutionary method of Seader and Westerberg and also the second sequence during the evolutionary synthesis by the method of Nath and Motard . In Table 5, the performance of our method is seen to be more efficient than previous techniques

Table 5: Comparison of various for example		
Method	$N_{sd}$	F %
Ordered branch search [25]	18	91
Predictor based ordered search [26]		50
Heuristic and evolutionary approach [8]		21
Evolutionary approach <sup>[7]</sup>	4	26
Ordered heuristic method [22]		14
Thermodynamic search algorithm [5]		12
Proposed method		

Table 5: Comparison of various for example 1

TESCE , Vol.29, No.2 -186- -186- July 2003

### *Example 2: Separation of a seven-component mixture*

Consider the multicomponent separations involved in the large scale thermal cracking of hydrocarbons to ethylene and propylene [23]. The feed mixture is given in Table 6 .A summary of the results of the proposed method is shown in Table 7. Thus the resulting sequence is **[ABCD/EFG, AB/CD, EF/G, A/B, C/D, E/F].** The sequence is used by the industry in the thermal cracking of naphtha's [23] and is in good agreement with the second sequence obtained by Nadgir and Liu [22]. This result is the same obtained by Aly [24] using analogical gates

Component	Mole fraction	Normal boiling	Relative
		point difference (k)	volatility
A: Hydrogen	0.20		
		92	0.911
B: Methane	0.055		
		57	11.304
$C:$ Ethylene	0.267		
		16	1.549
$D:$ Ethane	0.167		
		40	3.379
E: Propylene	0.155		
		6	1.208
F: Propane	0.067		
		$\bullet$ 41	3.853
G: n-Butane	0.089		

Table 6: Example 2 data

#### *Example 3: Separation of an eight component mixture*

Let us consider the example given by Bezzina et al. [27]; it concerns the separation of an 8-component mixture explicit in Table 8. Summary of the results of the proposed method is shown in Table 9.The resulting sequence is **[ABCD/EFHG, AB/CD, EF/HG, A/B, C/D, E/F, H/F]** .This result is the same obtained by Bezzina et al. [27] using statistical approach, and by  $\text{Airy}[24]$ using analogical gates.

Component	Mole fraction	Normal boiling	Relative
		point difference	volatility
		(k)	
A: Methane	0.050		
		72.8	8.352
<b>B</b> : Ethane	0.050		
		40.9	4.038
C:Propylene	0.100		
		5.7	1.236
D:Propane	0.100		
		30.2	3.073
E: i-Butane	0.200		
		11.4	1.529
F: n-Butane	0.125		
		36.5	4.327
G :Pentane	0.208		
		32.7	4.152
H: Hexane	0.167		

Table 8: Example 3 data

# **7- Conclusions**

Starting from a purely heuristic procedure based on the evaluation of four rules and noting some conflicts between these rules, we have proposed a fuzzy rule based procedure. The proposed method when applied to problems previously reported in the literature yielded optimum solutions which are consistent with the reported values and it overcomes some of the drawbacks of using heuristic, evolutionary and mathematical programming. The proposed algorithm is characterized by its simplicity and can be implemented by hand calculations,

8-References

[1] J.E. Hendry, D.F. Rudd and J. D. Seader, Synthesis in the design of chemical process. *AIChEJ* 19, 1(1973)

[2] V. Hlavacek, Synthesis in the design of chemical processes. *Computers chem. Engng* 2, 67(1978)

[3] N .G. Nishida, G. Stephanopoulos and A.W.Westerberg, A review of process synthesis,  $AIChEJ, 27,578(1981)$ 

[4] A. W. Westerberg, The Synthesis of distillation-based separation systems. *Computers chem. Engng* 9, 421(1985).

[5] M. A. Gomez and J.D. Seader, Synthesis of distillation trains by thermodynamic analysis, *Computers chem. Engng* 9, 311(1985)

[6] M. K. Garg., P. L. Dougles and J. G. Linders, An expert system for identifying separation processes. *Can. J. Chem. Eng.,* 69,67(1991)

[7] R. Nath and R.L. Motard, Evolutionary synthesis of separation processes, *AIChEJ* 27, 578(1981)

[8] J. D. Seader and A. W. Westerberg, A combined heuristic and evolutionary strategy for synthesis of simple separation sequences , *AIChEJ* 23, 951(1977)

[9] L.Pibouleau and S. Domenech, Discrete and continuous approaches to the optimal synthesis of distillation sequence, *Computers chem. Engng*  10,479(1986)

[10] J.E. Hendry and R.R. Hughes, Generating separation process flowsheets, *Chem. Eng. Prog* 68,71(1972)

[11] A. W. Westerberg and G. Stephanopoulos, Studies in process synthesis : part 1 branch-and-bound strategy with list techniques for the synthesis for separation schemes, *Chem Engng Sci.* 30, 963(1975).

[12] M. J. Andrecovich and A. W. Westerberg, A MILP formulation for heat-integrated distillation sequence synthesis, *AIChEJ*31, 1461(1977).

[13] L. A. Zadeh, Fuzzy sets, *Inform. Control.* 8,338(1965).

[14] D. Dubois and H. Prade, Fuzzy sets and systems, Academic Press, NewYork(1980).

[15] A. Kraslawski, Review of applications of various types of uncertainly in chemical engineering. *Chem. Engng. Process,* 26,185(1989).

[16] L. A. Zadeh, In Introduction to the theory of fuzzy subsets, Vol.1 (A. Kaufmann, Ed.) pp . ix-x. Academic Press, New York (1975).

[17] H. J. Zimmermann, Fuzzy set theory-and its application, Chap. 10, Kluwer-Nijhoff Publishing, Hingham MA (1985).

[18]S . Aly, S.Domenech, L. Pibouleau, and P. Fliquet, Heuristic approach for designing distillation sequences, *AMSE Periodicals Modeling Measurements & Control,* 36,1(1993).

[19] R. W. Thompson and C. J. King, Systematic synthesis of separation schemes, *AIChEJ*8, 941(1972).

[20] P. C. Wankat, Equilibrium staged operations, Elsevier, New York, (1988).

[21] T.L. Flowers, B.K. Harrison and M.J. Niccolai, Automated synthesis of distillation sequences using fuzzy logic and simulation, *AIChEJ* 40, 1341(1994).

[22] V. M. Nadgir and Y.A. Liu, Studies in chemical process design and synthesis: part V. A simple heuristic method for systematic synthesis of initial sequences for multicomponent separations, *AIChEJ* 29, 926(1983).

[23] G.J. King, Separation processes, 2<sup>nd</sup> edn. McGraw-Hill, New York, (1980).

[24] S.Aly, Fuzzy analogical gates for separation sequence synthesis, *Chem, Engng. Process* 36,209(1997)

[25] F.R. Rodrigo and J.D.Seader, Synthesis of separation sequences by ordered branch search,, *AIChEJ29,* 926(1983).

[26] M.A. Gomez and J.D.Seader, Separation sequence synthesis by a predictor based ordered search, *AIChEJ* 22, 970(1976).

[27] M. Bezzina, L. Piblouleau and S. Domenech, A statistical approach to the synthesis of separation sequences, *Computers chem. Engng* 13, 651(1989).

# **9-Appendix**

#### *Definition and Calculation of Estimated Mass Load* (EML) *Coefficients*

The estimated separation mass load (EML) is the molar flow rate that has to be processed by all separation units downstream of the current separator before all desired products are isolated [24]. The numerical value of EML is the weighted probability of each possible downstream sequence, without referring to the physical properties of components. The main result are listed in the following table (where  $x_i$  is the molar fraction of the component i in the mixture):



For each split, an EML coefficient can be derived; by application of a linear programming code, two bounds  $EML_{min}$  and  $EML_{max}$ can be computed .The following table shows these two bounds for some splits.





0.333

 $\frac{1}{1.333}$ 

 $\overline{0.333}$ 

 $1.000$ 

 $0.500$ 

 $\frac{100}{2}$ 

 $\frac{0.667}{ }$ 

 $\frac{1}{3.00}$ 

 $\overline{0.50}$ 

 $\frac{36.50}{ }$ 

DEF









0.287

0.287

0.287

1.000

 $0.403$ 

1.000

0.713

 $3.853$ 

 $\frac{1}{0.403}$ 

 $\ddot{+}$ 

EFIG



Table 9: Summary of the results of example3 Table 9: Summary of the results of example3

TESCE, Vol.29, No.2