CASE-BASED REASONING FOR SEPARATION PROCESS SYNTHESIS

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ABSTRACT

A method of reusing existing design cases for synthesis of distillation process sequences is presented. The approach uses case-based reasoning (CBR) that finds the most similar existing separation designs and applies the knowledge of their concept for solving new problems. The method has previously been developed for selection of single separations and simple sequences but has now been extended to cover synthesis of more complicated sequences. The method is intended for finding feasible process alternatives in preliminary process design.

INTRODUCTION

The paper presents a method for finding feasible separation process sequences by using case-based reasoning (CBR). CBR is a method of reusing existing design cases for making new designs. This means finding most alike existing processes and applying the knowledge of their separation capacity and design for solving new design problems. This is especially important in the early phases of process design when many alternatives should be quickly screened before a more detailed study is done. There is a great need for these kinds of screening tools to reduce the number of design options and quicken the process design in practice [1].

When dealing with multicomponent mixtures, the number of possible separation methods, their combinations and process structures to be screened is huge as well as the work involved. The problem of synthesis of a simple separation sequences and the selection of single separations has been studied earlier [2, 3, 4]. However a comprehensive CBR-based methodology for the synthesis of more complicated separations sequences has not been presented yet.

CBR IN PROCESS SYNTHESIS

Case-based reasoning (CBR) is one of the non-symbolic AI methods [5]. CBR solves new problems by finding and adapting existing successful designs for solving new problems (Fig.1).

The lack of systematic reuse of existing design experience has been a shortage in process design. The main benefit of CBR approach is that readily available existing knowledge can be utilised systematically also in large and complex problems such as process synthesis and design. In this way the time-consuming conceptual screening phase of a design project can be fastened. CBR is attracting attention, because it seems to directly address the conceptual process design problems outlined earlier.

Some benefits of the CBR approach are:

1. CBR does not require an explicit domain model and so elicitation becomes a task of gathering case histories.
2. Implementation is reduced to identifying significant features that describe a case, which is an easier task than creating an explicit model.
3. By applying database techniques, large volumes of information can be managed.
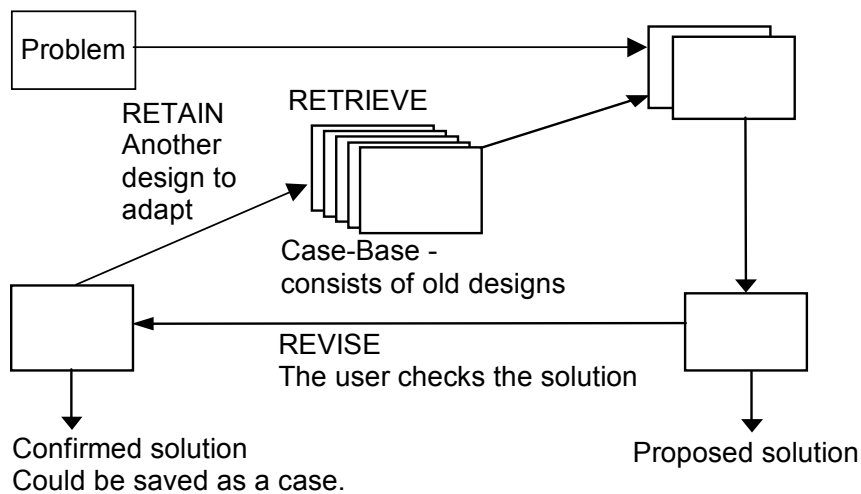4. CBR systems can learn by acquiring new knowledge as cases, thus making maintenance easier.

Figure 1. Principle of case-based reasoning

Because generalisations are not needed in CBR, no data is lost. CBR gives answers to design problems in a straightforward way. The results are dependent on the retrieval parameters and the adaptation applied. The strong interaction with the user makes the flexible and interactive use of existing data and design experience possible. The CBR search can be focused on different aspects by defining new search criteria and weighting retrieval criteria differently. In this way the same case base can be used for several types of tasks. The system learns by updating the information of the database.
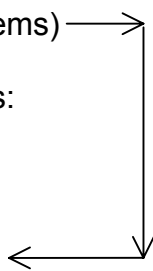
CASE RETRIEVAL

A retrieval algorithm using the indices in the case-memory should retrieve the most similar cases to the current problem or situation. The retrieval algorithm relies on the indices and the organisation of the memory to direct the search to potentially useful cases. The issue of choosing the best matching case has been addressed by research on analogy. This approach involves using heuristics to constrain and direct the search [5].

Methods for case retrieval are nearest neighbour, induction, and knowledge-guided induction and template retrieval. These methods can be used alone or combined into hybrid retrieval strategies. If the nearest neighbour is used, then case features should be able to be weighted and similarity measures customised. If inductive techniques are used, the index tree generated should be open to inspection and alteration by developers. A typical equation for calculating nearest neighbour matching is Eq. 1, where w is the importance weighting of a feature, sim is the similarity function, f^I and f^R are the values for feature i in the input and retrieved cases respectively [5].

$$\frac{\sum_{i=1}^{n} w_i \times sim\left(f_i^I, f_i^R\right)}{\sum_{i=1}^{n} w_i} \tag{1}$$

THE CBR-BASED SEPARATION SYNTHESIS ALGORITHM

The main phases of general CBR-based separation process synthesis algorithm consist of 1) selection of the methods of single separations, 2) selection of separation sequences and 3) selection of combined (hybrid) separations. The phases of the algorithm are listed below and discussed in the following sections in more detail:

1. Selection of single separations
   a) Search for the feasibility of conventional distillation based operations
   b) Search for azeotropes (see subcase; synthesis of azeotropic systems)
   c) Search for suitable mass separation agents (MSA)
   d) Search for other (non-conventional distillation) separation methods:
      i)   calculation of relative physical properties (R's)
      ii) search for separations based on feasible relative properties

   1B. Subcase; selection of azeotropic separations
       a) Search for separation in column in isobaric conditions
       b) Search for separation in columns in non-isobaric conditions
       c) Separation by using MSA
       d) Separation by using MSA and non-isobaric pressure
       e) Separation by other means; reactive, membrane, extraction etc.
       f) Separation by hybrid or combined operations

2. Separation sequencing by using as search criteria:
   - component names or types

- relative volatilities of components
- VF values of components
- CES values of components

And applying:

a) *sequences* in found cases *or*
b) sequence *heuristics* (if they are stored with cases)

3. Search for combined separation operations

SELECTION OF SINGLE SEPARATIONS

The feasibility of ordinary distillation is studied first in the methodology, since it is the most common way of separating fluid mixtures. This is done by studying the distillation related properties (relative volatilities and lacking chemical reactivities) in the first step. The remaining separation problems are solved with further reasoning, which applies separation methods other than ordinary distillation. In this phase relative properties are calculated and the values that show potential for separation are used as retrieval parameters. The main steps of the approach of selecting single separations are [2]:

Step 1: Conventional distillation is usually applied whenever the relative volatility (α) is large enough. The first search for the solution is made using component names, α's and reactivities as retrieval parameters. To make the search simpler, α's can be classified as easy ($\alpha \geq 1.2$), possible, where mass separating agent (MSA) could be useful ($1.1 < \alpha < 1.2$) and difficult ($\alpha \leq 1.1$). A more accurate search is made (capacity and component types also as retrieval parameters), if several alternatives are found. The nearest strategy found is then applied in all the separations, where ordinary distillation is applicable.

Step 2: To be able to compare separation methods, where mass separating agent (MSA) is needed, suitable MSA is searched for each binary component pair that cannot be separated by conventional distillation. The retrieval parameters used are e.g. the types of components, concentrations, relative solubility parameter, dipole moment and dielectric constant. The found MSA is used for defining solubilities and other separation related properties in step 3.

Step 3: Relative physical property parameters [6] are calculated for each component pair that can't be separated by ordinary distillation. The parameter values are compared to the feasibility limits of different separation methods.

Step 4. Separation methods are searched using the relative parameters (min and max values), that are within the feasibility limits as retrieval parameters. For example crystallisation is considered very feasible if the relative melting point is greater or equal to 1.2. Also a more detailed search by using concentration, capacity and component types as retrieval parameters can be defined.

The possibility of combined operations should be checked. This is done in the last phase of the main algorithm and discussed later.

SYNTHESIS OF AZEOTROPIC SEPARATIONS

The presence of azeotropes adds some difficulties to separations and also the synthesis problem becomes much more complex. Therefore the synthesis of azeotropic distillations is a special case of the general synthesis algorithm.

In general to separate azeotropic mixtures various technologies may be used [7]:

1. Pressure-swing distillation. A series of column operating at different pressures are used to separate binary azeotropes, which change appreciably in composition over a pressure range or where a separating agent, which forms a pressure-sensitive azeotrope, is added to separate a pressure-insensitive azeotrope.
2. In homogeneous azeotropic distillation, a third component is added to modify the components relative volatility.
3. Heterogeneous azeotropic distillation is based on the same principle as homogeneous azeotropic distillation, but the added third component is partially miscible with one of the components, Solvent reprocessing is easy by means of a liquid-liquid separation system.
4. Reactive distillation is based on the transformation of one of the components into a component, which does not form an azeotrope with the other components.
5. Salted distillation consists in adding an ionic salt that dissociates in the liquid mixture and changes the azeotrope composition.
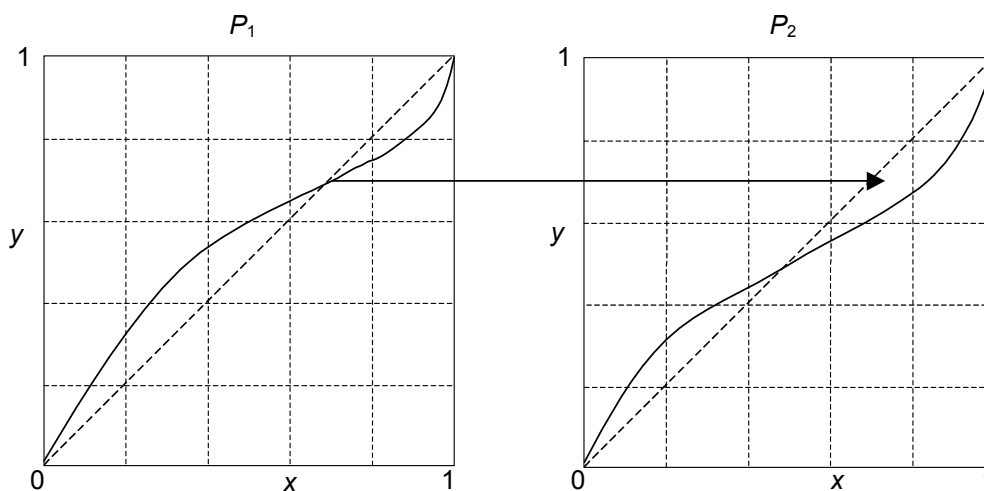

Fig. 2 y-x –diagrams at different pressure ($P_1 < P_2$) for minimum-boiling azeotrope
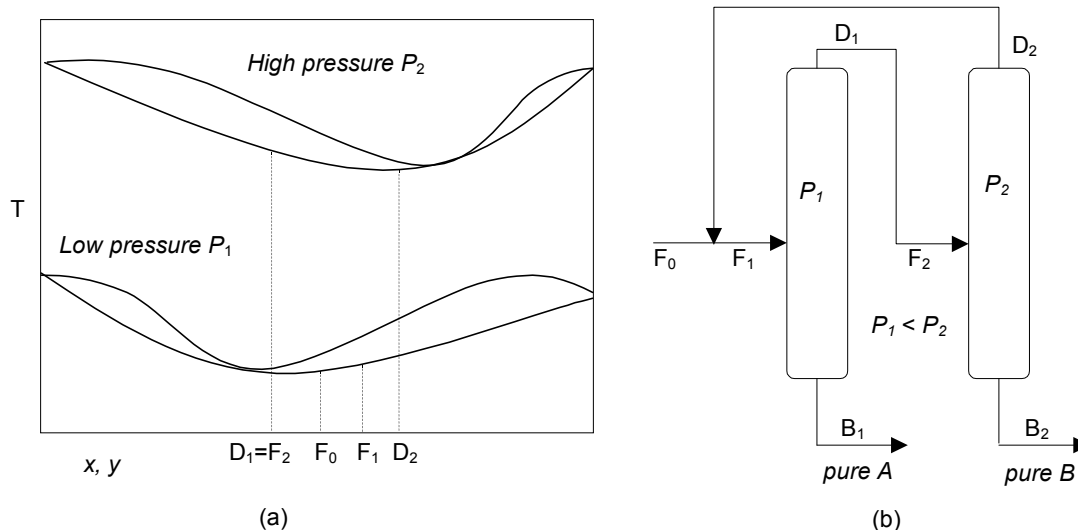
Fig. 3 Pressure-swing distillation: (a) temperature-composition diagram for a minimum-boiling binary azeotrope that is sensitive to changes in pressure; (b) distillation sequence.

Pressure changes can have a large effect on the vapor-liquid equilibrium compositions of azeotropic mixtures and thereby affect the possibilities to separate the mixture by ordinary distillation. By increasing or decreasing operating pressures in individual columns the distillation boundaries can be moved in the composition space or the azeotropes can even be made to appear or disappear. (Fig. 2 and 3).

The synthesis algorithm for azeotropic separations is a modification of the general synthesis algorithm [4]. The hierarchy of searches in the algorithm is the following:

1. Separation in single or multiple columns in isobaric and non-isobaric pressure
2. Separation by using MSA
3. Separation by using MSA and non-isobaric pressure
4. Separation by other means; reactive, membrane, extraction etc.
5. Separation by hybrid or combined operations

When azeotropes are present in the mixture, the definition of case description and retrieval parameters is more complex. One idea is to use the relative similarity based on the similarity of feed, product and azeotropic points. To be able to compare separation methods, where mass separating agent (MSA) is needed, a suitable MSA is searched for each binary component pair that cannot be separated by conventional distillation. The first retrieval parameters used are types of components to be separated. Also a more accurate search is defined (concentrations, relative solubility parameter, polarity and dielectric constant as retrieval parameters), if several alternatives are found. The found MSA is used for defining solubilities and other separation related properties. If the MSA has not been used earlier for same components, more rigorous studies, simulations and/or experiments are needed to confirm the suitability. An example on searching a MSA is given later.

EXAMPLES ON SINGLE SEPARATIONS

Separation of Azeotropic Solution

As an example on single separations the separation of pyridine from water is studied as an example. Since the separation concept is determined by the concentration of product streams required, the composition of the azeotropic point and the solubility of the mixture (i.e. if there is a phase split), these are also the search criteria used in CBR. If the criteria are analogous to the case found in database, the process concepts are similar too and the concept found can be reused.

Problem: Dilute pyridine water solution needs to be separated into products containing 40 and 1 wt-% of pyridine.

Search criteria:
1) Azeotropic composition of pyridine with water: 94 °C and 57 wt-%
2) Solubility with water: Total
3) Feed composition: 15 wt-% pyridine
4) Product compositions: 40 and 1 wt-% pyridine

Using these parameters the nearest case found is:

Components: Tetrahydrofuran (THF) and water
Azeotrope: at 64 °C 96 wt-% THF
Solubility: Total (at 70 °C)
Feed: 20 wt-% THF
Products: 55 wt-% and 3 wt-% THF
Separation: Distillation in a single column without entrainer

The found case is analogous in the azeotropic behaviour and in relative stream concentrations (especially relative to the azeotropic point). In neither cases the azeotropic concentration is crossed. Based on this, it can be reasoned that distillation in a single column is applicable also in the pyridine case searched and no entrainer is required.

Selection of Mass Transfer Agent (MSA)

Problem: Find suitable mass separation agent for THF/water separation.

The search is made using following retrieval parameters: component type, solubility parameter, dipole moment and dielectric constant (these three describe solvent's separation capability). The nearest cases are shown in Table 1.

Table 1. Query and nearest cases in the THF/water problem

	Query	Found 1	Found 2
Component 1 type	Water	Water	Water
Component 2 type	Ether	Ether	Acetate
Component 1	Water	Water	Water
Component 2	THF	Diethyl ether	Ethyl acetate
Solubility parameter	9.9	7.4	9.1
Dipole moment / D	1.75	1.3	1.7
Dielectric constant	7.6	4.34	6.02
MSA's		n-Hexane, Benzene, Toluene	n-Pentane, 2,2-Dimethyl-butane, Dichloromethane
Similarity		0.92	0.85

The found MSA is used for defining solubilities and other separation related properties for step 3. If the MSA has not been used earlier for exactly the same components, more rigorous studies, simulations and/or experiments are needed to confirm the suitability. In this case n-pentane has been reported for THF/water separation [8, p.108].

Separation in Non-Isobaric Distillation System

Problem: Separate tetrahydrofuran (15 wt-%) from water. Purity requirement for tetrahydrofuran product is 99 wt-%.

This cannot be reached with ordinary distillation, because tetrahydrofuran and water form an azeotrope at 64 °C with 96 wt-% THF [8, p. 328].

Search criteria:
1) Feed composition: 15 wt-% THF
2) Product compositions: 99 wt-% THF
3) Use pressure-swing distillation
4) One or more columns
5) No MSA

Using these parameters the nearest cases are shown in Table 2.

Table 2. Query and nearest cases for non-isobaric separation

	Query	Found 1	Found 2
Component 1	Water	Water	Water
Component 2	THF	THF	THF
Feed	15 wt-%	10 wt-%	12 wt-%
Products	99 wt-% THF	99 and 3 wt-% THF	95 and 4 wt-% THF
Nr. of columns	-	2	1
Comments	-	Distillation in a two column system operating at different pressure (p_1=1 bar, p_2=7,6 bar)	Distillation in a one column system operating at different pressure (top1 bar, bottom 8,0 bar)
Similarity		0.95	0.85

It is found that the THF / water system can be separated by shifting the azeotropic concentration by changing the system pressure in one or two column systems (Fig. 4).
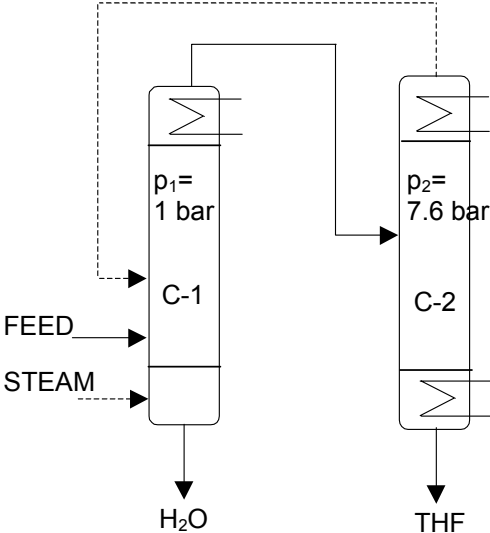

Fig. 4 Separation of THF / water mixture in two columns system.

FINDING SEPARATION SEQUENCES

Alternative approaches in selection of separation sequence

In the synthesis approach presented the selection of the separation sequence starts from choosing of a single separation as shown before. There are several alternatives for the selection of a separation sequence:

1) Finding all possible separation sequence combinations. This is feasible only in small cases, because the combinatorial explosion takes place quickly when the number of products to be separated increases.

2) Using optimisation algorithm such as or genetic algorithms (GA) [9] or mixed integer programming (MINLP) to find the most feasible separation sequence. Both approaches require the selection of a superstructure. Other disadvantage is the limitation on the size of the problem handled because of the combinatorial explosion, which is problematic especially with MINLP.

3) The case-based reasoning approach can be used through an 'upper level' CBR. This is discussed in more detail in the following section.

Separation sequence synthesis by CBR

The case-based reasoning approach can be used through an 'upper level' CBR for finding out a separation sequence. This is possible since a database can also be used for storing feasible separation sequences - not only information on single separations.

The search can be done:
i)   directly with *component names* or
ii)   *component types* (e.g. aliphatic alcohol) or
iii)   in a more creative way by using analogies through *characteristic properties* of the components to be separated.

There are two alternative ways to interpret the search results:
i)   directly as feasible separation *sequences* or
ii)   as feasible separation *sequence heuristics*, which can to be applied on the design of the new sequence. In this approach the heuristics are stored together with the cases and the sequence is adapted from the strategy of the nearest existing case found [2].

In the first sequencing strategy it is possible to make searches not by names but criteria, which are related to the separation properties of the component pairs. The criteria should describe the difficulty of the separation by using properties such as boiling points, relative volatilities or coefficients of ease of separation (CES) [12, p. 154]. However, in the end we are interested in the costs of the required separation tasks. The cost of separation is not only dependent on the physical separation properties of the components but also on their concentrations in the feed and in the required products. Therefore the use of CES is a possible way of making searches. However the value of CES is dependent on the separation sequence and consequently its use as search criteria is difficult. Therefore a new more straightforward search criterion is developed in the following.

It is well known that the column operating and capital costs are related to the vapour flow of the column. Porter and Momoh [10] have suggested Eq.2 as an approximate

method of calculating the vapour flow V in a column, which can also serve as a simple estimate on the both costs:

$$V = D\left[1 + \frac{R_F}{(\alpha - 1)}\frac{F}{D}\right] \tag{2}$$

where

$V =$ vapor flow
$D =$ distillate flow rate
$F =$ feed flow rate
$\alpha =$ relative volatility

$$R_F = \frac{R}{R_{\min}} \tag{3}$$

$R =$ reflux rate
$R_{min} =$ minimum reflux rate

The Equations 2 and 3 can be simplified to form the search criterion VF, which is calculated for all the component pairs to be separated:

$$VF = (D + \frac{1.1F}{\alpha - 1})N \tag{4}$$

R_F was substituted with a typical value of 1.1. D and F are not the molar flows but percentage concentrations of the component pair. N is the number of components in the separation task. N is used for scaling to make the values comparable between different separation problems.

If other methods than a conventional distillation is used, the values of relative volatilities are scaled to give a correct view of the economic feasibility. For extraction and extractive distillation the method of Sounders [11] is employed for cost scaling. Other types of cost comparison charts are available elsewhere [12, p. 90].

The method of finding separation sequences by CBR is the following: After the single separation methods are determined, the VF values are calculated for all component pairs to be separated by using percentage concentrations in the feed of the whole system. The VF is scaled with the number of components in the feed N to make the values comparable with problems of unequal number of components. After this the search is made in the database, which has known separation sequences stored with the component VF values. Since several sequences are often nearly as good at least in the theoretical sense (i.e. they give nearly similar values for the objective function) [9], [12, p. 141], it is useful to store several feasible sequences for one problem into the database.

It is also possible to split the problem into subproblems and search for these subsequences. In this case a smaller part of the problem is searched from the database at a time.

The similarity (s) between values a and b is defined by Eq. 5:

$$s = 1 - \frac{|a - b|}{a}$$

(5)

The similarity (s) can be calculated based either on absolute or relative values of properties. For instance the VF values can be scaled to unity for a certain base component to calculate relative similarities. For absolute similarities there is no scaling. The similarity for the whole separation sequence is the average of the similarities of the single separations. The approach for separation sequence synthesis is demonstrated by the following sample problem.

EXAMPLE ON SEPARATION SEQUENCE

<u>Problem</u>. Separate the mixture of light hydrocarbons shown in Table 3 into 'pure' components.

Table 3. Problem feed compositions, adjacent relative volatilities and VF values

No.	component	mol-%	α	VF	corresponds
1	propane	13.7	2.53	224	y
2	i-butane	11.7	1.26	588	y
3	n-butane	5.5	2.39	126	y
4	i-pentane	9.9	1.30	1000	x
5	n-pentane	26.4	2.16	400	x
6	i-hexane	5.6	1.31	854	x
7	n-hexane	27.2			

The values of VF are calculated for adjacent separations and search is made in the database. The search can be made for similar (s_1) or relatively similar (s_2) sequences.

Table 4. Found feed compositions, adjacent relative volatilities, VF values and similarities

No.	component	mol-%	α	VF	s_1	s_2	corresponds
1	A	57.6	1.47	820	0.82	0.5	x
2	B	5.5	1.72	164	0.41	1	x
3	C	17.7	1,42	456	0.53	0.75	x
4	D	19.2					
average similarity					0.59	0.63	
1	Benzene	31.0	1.90	545			-
2	Toluene	33.0	1.76	450	0.50	0.74	y
3	EB	6.6	1.06	2930	0.20	0.54	y
4	Xylenes	25.0	1.76	340	0.37	1	y
5	C9s	4.9					
average similarity					0.35	0.64	

The most similar sequences found are shown in the Table 4 with corresponding subsequences in Table 3 marked with x and y. It can be seen that both the relative (s_2)

and absolute similarities (s_1) are useful, since they reflect a different point of view to the problem (i.e. relative or absolute resemblance).

The sequence synthesised based on the combination of the two found best cases [13, p.180], [14, p.140] is shown in Figure 5. The shadowed separations are from the first case (x) and the non-shadowed from the latter case (y). Note that not all separations of the latter case were used. The synthesis result corresponds the optimum result reported [13, p. 218].
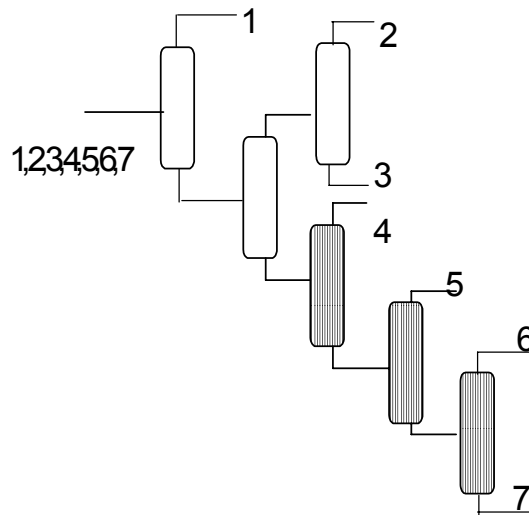

Figure 5. The synthesised separation sequence

COMBINED OPERATIONS

After the separation sequence synthesis the possibility of combined operations should be studied. For example a single column can separate several products using side streams. The approach for this is first to consider conventional separation sequences and then try to combine single separations one by one. Alternative approach is to conclude possible combination operations from the retrieved cases.

Example. Consider distillation for a four component mixture (A/B/C/D), where A is a very light component. The originally synthesised separation sequence is shown in Figure 6.
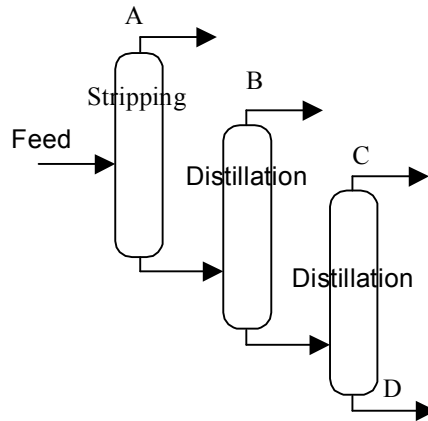
Figure 6. Train of single distillation separations

Searching for possible combination operations would give a case for hydrocarbon separations with uncondensable gases where uncondensables are taken out from the condenser as a third stream. Applying this case to the problem would give a combined system shown in Figure 7.
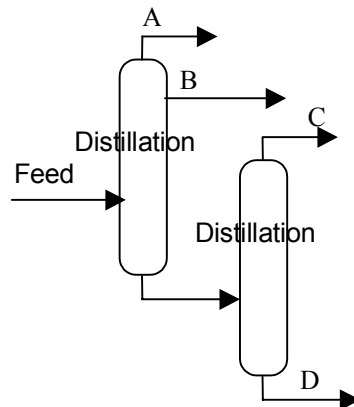

Figure 7. The train with a combined operation

CREATIVITY AND LEARNING ASPECTS

An important aspect in process design is creativity. A design system should not only be capable of modifying existing designs included in the database but also able to create new designs. One possible way of including creativity into synthesis is to use analogies [3]. Analogies can be included by using 'generalisations' and structural features such as proper hierarchy. The generalisations introduced may include general level categories in database such as type of separation, type of components or their physical properties.

CONCLUSIONS

The case-based reasoning method for separation process synthesis is applicable especially in conceptual process design for screening options to be studied by simulation in more detail. The advantage of CBR compared to rule-based methods is that all the existing knowledge is available as detailed cases and can be utilised in a non-reduced form. The method is flexible, since the user can extract various types of information even from the same cases by using different types of search criteria,

weights and similarity measures depending on the requirements and point of view. The method aims not to substitute the process designer or process simulation but the goal is to give a design tool to support the designer.

The method compares with the optimization based methods (e.g. MINLP) in the following ways: Optimization requires an explicit objective function to be defined. However many design criteria such as safety or operability are difficult or impossible to quantify and therefore include in the objective function. The optimization approaches are not interactive; the optimization cannot be guided easily by the user as in CBR. Therefore optimization often finds impractical or infeasible optima. The use of optimization requires that the optimization alternatives and area has to limited. This tedious task called 'generation of superstructure' has to be done by the user. If done improperly, it has a detrimental effect on the result. If the superstructure selected is too large, a combinatorial explosion will take place, if too narrow, feasible options are cut off.

The characteristic (and problem) of CBR is its dependency on the database and the engineering skills of the designer. On the other hand the method can utilise the huge amount of data available in the literature, if the information can only be extracted. Therefore methods of data mining become of importance. The general quality of design can be improved, since CBR enhances the systematic reuse of existing design experience, especially if the design cases are stored with the feedback and practical experience gained from existing engineering designs. Ultimately the database generated can form a valuable 'institutional memory' of the company.

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