Selection of separation sequences by case-based reasoning

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The objective of this paper is to introduce a method for finding feasible separation process sequences and separation process structures utilising case-based reasoning (CBR). This means finding the most similar existing separation processes and applying the knowledge of their concept and separation sequencing for solving new problems in the early phases of process design.

1. INTRODUCTION

Typical task in process design is to determine the configuration of a separation sequence. 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 method used, case-based reasoning (CBR), uses existing design cases stored in the database for solving new separation problems. The synthesis method studies the physical and chemical properties of the species present in the mixture and uses the properties presenting most favourable possibilities for successful separation for retrieving the nearest cases to the current problem. The presented method has the advantage of not losing any information because no generalisations are used.

2. METHODOLOGY

It is a well-known fact that in the majority of cases distillation is the most feasible way to separate components (Barnicki and Fair 1990). Therefore the distillation related properties are studied first in the methodology. The strategy is to find first a feasible distillation sequence for the separations where ordinary distillation is possible, and then to solve the remaining separation problems with further reasoning which apply other separation methods than ordinary distillation (steps 2-4 in Table 1). The main steps of this algorithm are presented in Table 1.

Step 1: For all components α's are calculated and the presence of reactive components is also considered. The most similar cases to the current problem are searched from database based on these parameters. When it is possible (and no known cases for better procedures are found) ordinary distillation is applied using the same separation strategy as defined by the sequence of the nearest case found in database. The separation strategy is described in the cases as a set of heuristic rules or as textual description.

The separations are classified in the database based on relative volatility (α) values as easy (α>=1.2), possible, where mass separating agent (MSA) could be useful (1.1< α <1.2) and difficult (α =<1.1) separations. In a simple situation a search using component names would

be the most exact, but in a case where exactly the same components are not found, a more advanced approach is more useful (see Chapter 3). If the cases found are equally similar to the current problem, the query should be made more exact using concentration, capacity, component types etc. also as retrieval parameters. Unfortunately a separation problem is often far more complicated than finding only a distillation sequence and also other separation methods than ordinary distillation have to be considered.

Step 2: To compare other separation techniques with mass separation agent aided operations we need to select a suitable MSA for each component pair if possible. This has been done using component names or component types as retrieval parameters. Component types are defined as a taxonomy tree (see Figure 1). The closer the components are in the tree the greater similarity value they have. The principle of similarity is discussed in earlier paper (Pajula 2000). Even if a promising MSA is found, also other separation methods are checked. If there is no proven case for certain MSA, a more detailed study should be done by computer simulations or experimentally (See Example 3.1). Alternatively MSA can be searched using solubility parameter, dipole moment and dielectric constant as retrieval parameters.

Table 1. The algorithm

Step 1	DISTILLATION FEASIBILITY *Search:* Make a search with α's and reactivities as retrieval parameters *Refine*: Define a more accurate search (capacity and component types also as retrieval parameters) if several alternatives are found. *Action:* Apply the separation strategy of the nearest case for the separations where ordinary distillation is possible. If ordinary distillation is not feasible for all separations, continue to step 2.
Step 2	FINDING A SUITABLE MASS SEPARATION AGENT (MSA) *Search:* Make a search with component types as retrieval parameters for each remaining component pair. *Refine*: Define a more accurate search (concentrations, relative solubility parameter, polarity and dielectric constant also as retrieval parameters) if several alternatives are found. *Action:* Use the found MSA (if any) for defining solubilities etc. for step 3.
Step 3	FINDING PARAMETERS FOR ALTERNATIVE SEPARATION METHODS *Action:* Calculate relative physical property parameters for each component pair that can't be separated by ordinary distillation and compare them to the feasibility limits of different separation methods.
Step 4	SEARCH FOR A SUITABLE SEPARATION METHOD AND STRATEGY *Search:* Make a search using the relative parameters (min and max values) that are within the feasibility limits. *Refine*: Define a more accurate search (concentration, capacity and component types also as retrieval parameters) if several alternatives are found. If there are still several alternatives left, make economical comparison. *Action:* Apply the separation strategy of the nearest case to the components that can't be separated by distillation.

Step 3: It is often important to consider also other separation methods than ordinary distillation. Therefore it is necessary to consider all the possible properties that may be utilised in separation processes and make a search based on these. The principle is to apply separation method that utilises the largest property difference of the components to be

separated. To do this relative properties are calculated for component pairs and compared to predefined feasibility limits (Jaksland et al. 1995, Qian and Lien 1994). For example crystallisation is considered very feasible if the relative melting point is greater or equal to 1.2 and feasible if it has a value between 1.1 and 1.2. The approach is used for finding the most important retrieval parameters for CBR, i.e. the parameters that show greatest potential for separation of the species that have too small α's for ordinary distillation. In this way the amount of retrieval parameters is limited to essential ones.

Step 4: The search is made using the relative parameter values of step 3 that are within the feasibility limits as retrieval parameters. The separation method or strategy of nearest found case is applied.

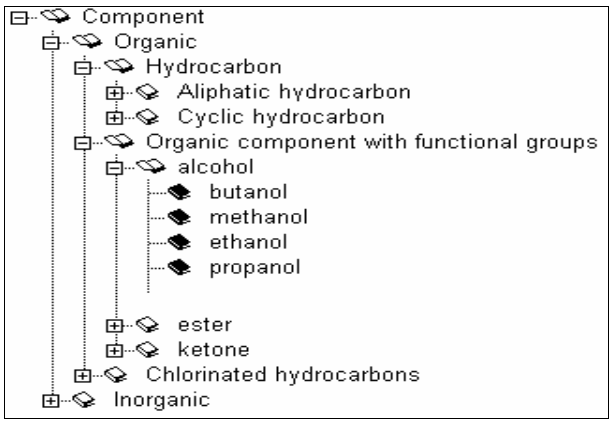

Figure 1. Part of the component type taxonomy

3.EXAMPLES FOR SINGLE SEPARATIONS

Example 3.1: Selection of mass transfer agent

In this example finding potential mass transfer agents for comparisons is demonstrated.

Task: Separate n-propanol (50 wt-%) from water using MSA. Purity requirement for n-propanol product is 90 wt-%. This cannot be reached with ordinary distillation, because water and n-propanol form an azeotrope at 87 °C with 71 wt-% n-propanol (Smallwood, 1993).

Table 2. Query and closest cases in Example 3.1

	Query	Found 1	Found 2
Component 1 type	Water	Water	Water
Component 2 type	Aliphatic alcohol	Aliphatic alcohol	Aliphatic alcohol
MSA	Yes	Yes	Yes
Component 1		Water	Water
Component 2		isopropanol	sec-butanol
Solubility parameter	11.9	11.5	10.8
Dipole moment / D	1.7	1.66	1.7
Dielectric constant	20.1	18.3	-
MSA's		Cyclohexane, Toluene, Diisopropylether, Isopropyl acetate	Butylacetate, Diisobutene, Benzene
Similarity	1	0.97	0.93

The first search is made using search parameters MSA=yes and component types (water, aliphatic alcohol). This gives quite a few cases with similarity value 1. In this case a more specific search is needed to find the most likely suitable MSA. The following additional retrieval parameters for alcohols are used: solubility parameter, dipole moment and dielectric constant. The two closest alcohols found and the MSA's used with them are summarised in Table 2. This is a realistic result for further studies, because at least benzene, diisobutene, diisopropylether and cyclohexane have been reported for n-propanol/water separation (Smallwood, 1993).

Example 3.2: Finding other solvent to replace current mass transfer agent

In this example finding an alternative for a MSA by utilising a case base that includes also component properties is demonstrated. Dimethylformamide (DMF) has been separated from water using chloroform as MSA, but other possible solvents are searched.

Task: Find a solvent the properties of which are close to chloroform and which is easy to separate from DMF.

The search is made using following retrieval parameters: solubility parameter, dipole moment, dielectric constant (these describe solvent's separation capability), solubility in water and solubility of water (the phases should be practically immiscible). Also boiling point and possible azeotrope with water (azeotrope boiling point) are used as retrieval parameters. This is important because the solvent has been separated from DMF (boiling point 153 °C) by ordinary distillation and the distillation column temperature should be high enough so that the temperature of cooling water is cold enough for condensation. The results are presented in Table 3. This is a realistic result because both methylene dichloride and carbon tetrachloride have been reported as MSA in DMF/water separations.

Table 3. Query and closest cases in Example 3.2

MSA	Query	Case1	Case2
Name	Chloroform	Carbon tetrachloride	Methylene dichloride
Boiling point °C	61	76	40
Solubility param.	9.3	8.6	9.7
Dipole moment	1.1	0	1.8
Solubility in water	0.82	0.077	0.077
Solubility of water	0.2	0.008	0.008
Dielectric constant	4.806	2.24	9.1
Azeotropes			
Water wt-%	97	96	99
B.p. °C	56	66	38
Similarity		0.95	0.92

4. EXAMPLE FOR FINDING SEPARATION SEQUENCE

Example 4.1. Separation sequence selection – xylenes

In this example the method is applied for finding alternative separation sequences when ordinary distillation is not suitable for all separations. To demonstrate the approach, the search is not done by component names but by relative properties.

Task: Separate mixture of ethyl benzene (20 wt-%), m-xylene (40 wt-%), o-xylene (20 wt-%) and p-xylene (20 wt-%) to pure products.

Step 1: By searching with α's and reactivities it is found out that ethyl benzene and o-xylene can be separated by ordinary distillation. The strategies used in the two nearest cases found (similarity 1) are:
Heuristics 1: Perform difficult separation last and favour 50/50 split.
Heuristics 2: Perform difficult separation last and use CES for finding the distillation order.
We choose to use CES to define the separation order for these separations, because the other heuristics found doesn't give answer for the separation order in our problem. According to CES, first o-xylene and then ethyl benzene are separated by distillation. For m-xylene/p-xylene separation the available α is too small for ordinary distillation.
Step 2: To find out possible MSA aided separation methods for this separation, we'd like to check for potential MSA as presented in Example 3.1. In search no feasible MSA's were found for a case where both components are aromatic and have low polarity.
Step 3: For finding other separation methods for m/p-xylenes reasonable retrieval parameters are needed. This means the relative parameters that are large enough to make a separation possible. The potential parameters found are relative melting point ratio and relative kinetic diameter. The calculated values for these are presented in Table 4.

Table 4. Some properties and calculated relative properties for Example 4.1

	T (boiling)/K	***T (melting)/K***	***CES*** *(1st separation)*	***Binary pair***	***R (melting point)***
ethyl benzene	409.35	178.2	0.54	p-xylene/m-xylene	1.27
p-xylene	411.51	286.4	0.506667		***R (kinetic diameter)***
m-xylene	412.27	225.3	1.3275	m-xylene/p-xylene	1.16
o-xylene	417.58	249.0			

Step 4: The database was searched using these relative parameters (R's) for melting point and kinetic diameter and only including cases in which at least one α-value is classified difficult. The results are presented in Table 5 below.
Based on the search results two feasible methods are proposed for further research: First separate o-xylene and then ethyl benzene by distillation. After this separate remaining p- and m-xylenes either by crystallisation or molecular sieve adsorption since these methods were found to be potential in the case-base search.

Table 5. Search results of Example 4.1

	Query	Found 1	Found 2	Found 3	Found 4
α's	difficult	difficult , easy	difficult , easy	difficult	difficult
R melting point. max	1.26	1.26	1.26	1.3	1.66
R kinetic diameter max	1.16	1.16	1.16		
Components		xylenes	xylenes	p- and m-dichlorobenzene	THF and water
Separation method		Distillation & molecular sieve adsorpt.	Crystallisation (only p-xylene separated)	Crystallisation	Molecular sieve adsorption
Similarity		1	1	0.8	0.7

5. OTHER ASPECTS OF THE METHOD

In these examples no combined operations were included in the case base. These are discussed in an earlier paper (Pajula et al. 2000). As separation methods, for instance hybrid membrane/distillation processes develop further, the case base needs to be updated. This can be done by adding rules or new cases that have low maturity. For instance, if the components can be separated by distillation, the heuristics presented by Rong et al. (2000) are notified. An other way is to create new cases by simulation. For instance, if the mole fractions (as retrieval parameters) in the feed stream are close to those where distillation the mole fractions in the feed are typical values for a complex distillation flowsheet, the case suggesting complex distillation flowsheet is retrieved among other near cases. When creating this kind of cases including new separation methods also process maturity factors (Pajula et al. 2000) and feasibility limits need careful attention.

6. CONCLUSIONS

A method for finding feasible separation process sequences and separation process structures utilising earlier design cases was developed. The demonstrations show how earlier design cases can help in selecting process alternatives to be considered especially at the early stages of a design and in this way fasten the design process. Also importance of updating the presented method was considered. The advantage compared to rule-based methods is that all the existing knowledge is available as cases and can be utilised in a non-reduced form. The method is also very flexible because the user can focus the search by defining more accurate search parameters if several nearly similar solution possibilities are available.

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NOMENCLATURE

α	relative volatility	μ	degree of feasibility (range 0-1)
R	relative parameter	R_{max}	maximum value of relative parameter