12TH INTERNATIONAL SYMPOSIUM ON PROCESS SYSTEMS ENGINEERING AND 25TH EUROPEAN SYMPOSIUM ON COMPUTER AIDED PROCESS ENGINEERING

PART B

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Alternative Hybrid Liquid-Liquid and Distillation Sequences for the Biobutanol Separation

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Abstract

Recent technologies for biobutanol production by fermentation have resulted in higher final biobutanol concentrations, less fermentation by-products and higher volumetric productivities during fermentation, together with less energy intensive separation and purification techniques. These new technology developments have the potential to provide a production process for biobutanol that is economically viable in comparison to the petrochemical pathway for butanol production. This study compares four different possible process designs for purification of biobutanol by means of a multiobjective optimization process having two objective functions: the total annual cost (TAC) as an economical function and the associated eco-indicator 99 as an environmental function.

Keywords: biofuels, biobutanol, thermal coupling, process synthesis.

1. Introduction

With the depletion in petroleum resources, an increasing attention has been paid to the research of new energy sources. The use of biofuels for transport is becoming very relevant for a number of reasons, such as environmental concerns relating to climate change, depleting fossil fuel reserves, and reducing reliance on imports (Wingren et al., 2003). This is leading to international, national and regional focus on alternative energy sources. In the EU, the transport is responsible for an estimated 21 % of all greenhouse gas emissions (Searchinger et al., 2008). More than 90 % of the total transport emissions are due to the road transport. Biofuels, processed from biomasses, which are considered as a renewable resource, are suggested as a direct substitute for fossil fuels in transport. Thus, the current research and development drivers are the identification of potential renewable energy sources or biomass feedstock and their processing in order to produce alternatives to fossil fuels in transport, such as biobutanol.
The aim of this work is to perform the process design, multiobjective optimization and comparison of different possible process routes for industrial scale biobutanol production. Process modelling in Aspen Plus was performed and the optimization was conducted using the stochastic optimization method Differential Evolution with Tabu list, having two objective functions, the total annual cost as an economic objective,

whereas the environmental objective function is measured through the eco-indicator 99 based on the LCA methodology.

2. Hybrid extraction-distillation process

The combination of extraction and distillation is considered as one of the most promising separation alternatives for the ABE purification (Van der Merwe et al., 2013). The extraction column is located after the fermentor, the mass separation agent or extractant, is fed from the bottom and the fermentation broth from the top. The raffinate phase contains water and traces of acetone, butanol and water. The extract phase contains the extractant, acetone, butanol and ethanol. The selection of the extractant is of meaningful importance for the economy of the process because directly affects the composition of the extract phase. It is possible to generalize that a good extractant, besides its economicity and low toxicity, should have a high distribution coefficient for the butanol and a high selectivity between butanol and water.
The extract phase obtained from the extractor is feed to the distillation section where acetone, butanol and ethanol can be recovered following different arrangements, with simple and/or complex columns. The liquid-liquid extraction may eliminate the request of azeotropic distillation making the process more competitive compare pure distillation flowsheets. The nine configurations reported in Figure 1 are considered and compared in the present study.

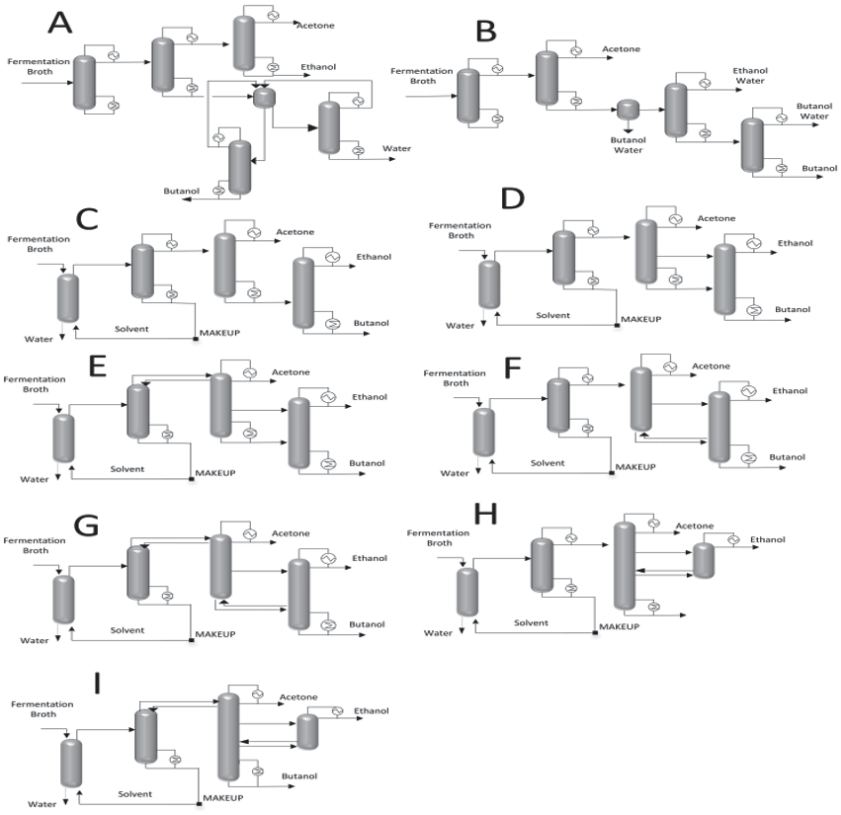

Figure 1. Sequences considered for the biobutanol production.

3. Case study

All sequences in Figure 1 were subject to a rigorous optimization, all these design cases were initially simulated using Aspen Plus process models. Note that these process models were robust and thermodynamically rigorous. According to Van der Merwe et al. (2013) and Chapeaux et al., NRTL-HOC was the most accurate thermodynamic model for the calculation of the physical property available for the components used at the specified conditions. It was assumed that all process designs have the same stream feeds except LLE design where it was added hexyl acetate as extractant. The feed composition reported by Wu et al. (2007) was used. The required purities are 99.5 wt % for biobutanol, 98 wt % for acetone, 94 wt % for ethanol and 95 wt % for the solvent recovered.

4. Optimization problem

The optimization of the biobutanol purification section is essential to run a biobutanol industry that can effectively compete with the current butanol derived from the petrochemical route. Moreover the environmental impact must be also taken in count in order to satisfy all governmental restriction.

4.1 Economic objective function

The objective function is the minimization of the total annual cost (TAC), which is directly proportional to the heat duty, services and the columns size. The minimization of this objective is subject to the required recoveries and purities in each product stream, i.e.:

$$\text{Min (TAC)} = f\,(N_{tn}, N_{fn}, R_{rn}, F_{rn}, F_{vn}, D_{cn}) \quad (1)$$
$$\text{Subject to } y_m \geq x_m$$

where N_{tn} are total column stages, N_{fn} is the feed stages of all streams in column, R_{rn} is the reflux ratio, F_{rn} is the distillate fluxes, F_{vn} is the vapor flow in interconnections at coupled sequences and D_{cn} is the column diameter, y_m and x_m are vectors of obtained and required purities for the m components, respectively. This minimization implies the manipulation of 22-26 continuous and discrete variables for each configuration, where 5-7 variables are used for the design of each column. Note that since the product streams flows are manipulated, the recoveries of the key components in each product stream must be included as a restriction for the optimization problem. In the configuration reported in Figure 1(a) the acetone, biobutanol and ethanol must be recovered; while in the configuration of Figure 1(b), the acetone and biobutanol must be recovered.

4.2 Environmental objective function

The environmental indicator is measured through the eco-indicator 99 based on the methodology of the life cycle analysis (Geodkoop and Spriensma, 2001) and is stated as follows:

$$\textit{Min (Eco indicator)} = \Sigma_b \Sigma_d \Sigma_k \delta_d \omega_d \beta_b \alpha(b,k)$$

Where β_b represents the total amount of chemical b released per unit of reference flow due to direct emissions $\alpha(b,k)$ is the damage caused in category k per unit of chemical b released to the environment, ω_d is a weighting factor for damage in category d, and δ_d is the normalization factor for damage of category d.
In the eco-indicator 99 methodology, 11 impact categories are considered:

1. Carcinogenic effects on humans
2. Respiratory effects on humans that are caused by organic substances
3. Respiratory effects on humans caused by inorganic substances
4. Damage to human health that is caused by climate change
5. Human health effects that are caused by ionizing radiations
6. Human health effects that are caused by ozone layer depletion
7. Damage to ecosystem quality that is caused by ecosystem toxic emissions
8. Damage to ecosystem quality that is caused by the combined effect of acidification and eutrophication
9. Damage to ecosystem quality that is caused by land occupation and land conversion
10. Damage to resources caused by the extraction of minerals
11. Damage to resources caused by extraction of fossil fuels.

These 11 categories are aggregated into three major damages categories: (1) human health, (2) ecosystem quality, and (3) resources depletion. These 11 categories are aggregated into three major damages categories: (1) human health, (2) ecosystem quality, and (3) resources depletion.

5. Global stochastic optimization strategy

In order to optimize the processes routes for biobutanol production, a stochastic optimization method, Differential Evolution with Tabu List (DETL) was used. Differential Evolution (DE) has its basis in Darwin's natural selection theory, and is similar to genetic algorithms (GAs) except for one important factor: several GAs, particularly earlier versions, encode decision variables as bit strings whereas DE encodes them as floating-point numbers. Srinivas and Rangaiah (2007) showed that the use of some concepts of the metaheuristic tabu can improve the performance of DE algorithm. In particular, the tabu list (TL) can be used to avoid the revisit of search space by keeping record of recently visited points, which can avoid unnecessary function evaluations. A comprehensive description of the DETL algorithm is provided by Srinivas and Rangaiah (2007).
The implementation of this optimization approach was made using a hybrid platform using Microsoft Excel and Aspen Plus. The vector of decision variables (i.e., the design variables) is sent to Microsoft Excel to Aspen Plus using DDE (Dynamic Data Exchange) through COM technology. In Microsoft Excel, these values are attributed to the process variables that Aspen Plus need. After simulation it is done, Aspen Plus return to Microsoft Excel the resulting vector. Finally, Microsoft Excel analyzes the values of the objective function and proposes new values of decision variables according to the stochastic optimization method used. For the optimization of process routes analyzed in this study, we have used the following parameters for DETL method: 200 individuals, 300 generations, a tabu list of 50 % of total individuals, a tabu radius of

0.0000025, 0.80 and 0.6 for crossover and mutation fractions, respectively. These parameters were obtained through a tuning process via preliminary calculations. The tuning process consists of performing several runs with different number of individuals and generations, in order to detect the best parameters to allow better convergence performance of DETL.

6. Results

In Figure 2 the results for the sequences of Figure 1(a) and 1(b) are reported. Only conventional columns are involved in separation and purifications of solvents. Pareto front from sequence 1(b) has a lower TAC and Eco 99 compare to the sequence 1(a), however it must be considered than in sequence 1(b), some part of biobutanol is wasted in the decanter, which means that recovery of biobutanol can not reach the target of 95 wt %.

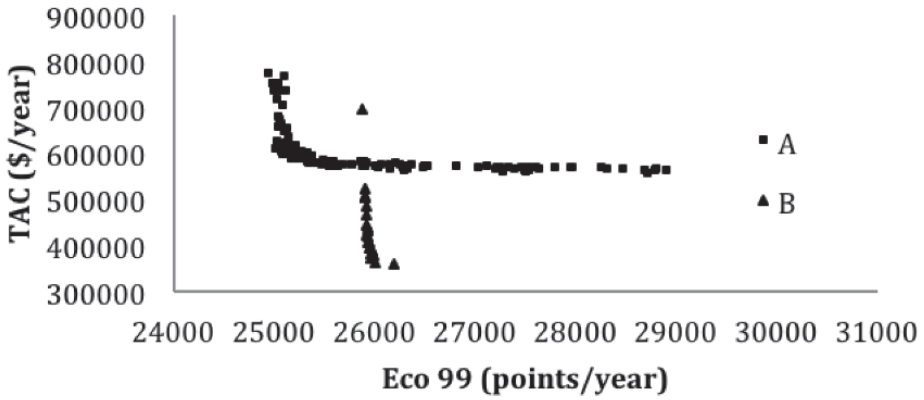

Figure 2. Pareto fronts from sequence of Figure 1(a) and 1(b).

In Table 1 the results of the TAC and the Eco-indicator 99 for all the alternatives considered, were reported. In general it is possible to notice that all the configurations with one or two thermal couplings are able to outperform the TAC and the Eco-indicator 99 of the classical configurations. Among all, configuration 1(h) reports the lowest value of TAC together with the second best environmental indicator.

Table 1: TAC and Eco 99 index for all the configurations of Figure 1.

Process Design	TAC ($/y)	Eco 99 (points/y)
A	580658	25351
B	363832	26009
C	128176	12975
D	133791	14974
E	104868	11056
F	123413	11499
G	107022	11729
H	101026	11434
I	108580	11168

7. Conclusions

A set of nine possible separation sequences for the biobutanol separation has been presented. Pure distillation alternatives together with liquid-liquid extraction plus distillation were considered and compared. The inclusion of thermal coupling in some liquid-liquid extraction based sequences reduced significantly the economic and environmental impact measured by TAC and Eco-indicator 99, comparing with some other sequences that do not include thermally coupling or a liquid-liquid extraction.
Compare to a pure distillation alternative, a liquid-liquid sequence with one thermal coupling was able to reach a 83 % reduction of the TAC together with 55 % reduction of the environmental indicator. This decreasing in economic and environmental issues, combined with other research efforts, could lead to be more profitable the use of biobutanol as an alternative energy source.

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