Research Article

Design and control of an alternative distillation sequence for bioethanol purification

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Abstract

BACKGROUND: Bioethanol is a green fuel considered to be a sustainable alternative to petro-derived gasoline. The transport sector contributes significantly to carbon dioxide emission and consequently has a negative impact on the air quality and is responsible for the increase of the greenhouse effect. The availability of environment-friendly and economical fuels is a worldwide priority. The separation process is a significant technology in the production of fuel grade ethanol in terms of both operating and capital costs. In the present work an alternative distillation sequence for bioethanol separation is presented. The steady state performance and the dynamic beavior are analyzed compared with the classical configuration reported in the literature.

RESULTS: Ethanol–water azeotropic separation represents a challenge for bioethanol purification. Usually a three column sequence is used to obtain fuel grade bioethanol by extractive distillation. In order to reduce bioethanol purification cost a two column separation sequence is proposed. This configuration shows a 10% saving in capital costs together with higher ethanol recovery and better control properties compared with the classical three column sequence.

CONCLUSIONS: Based on the steady state and dynamic results obtained, the two-column configuration represents a valid alternative to the classical configuration for the separation of bioethanol.
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INTRODUCTION

Problems concerning climate change, pollution prevention and diversification of energy sources are far from solved. At the present time, crude oil remains the most widely used energy source. World energy consumption has grown by about 60% in the last 30 years[1] and at the same time the International Energy Agency has estimated a decrease in crude oil production from 70 million barrels per day in 2007 to 27.1 barrels per day in 2030.[2] Focusing analysis on the transport sector, in Europe it accounts for 30% of total energy consumption, growing to 57.7% if the whole world is considered.[3] Usually oil derived fuels are used to cover this energy requirement increasing the dependence on politically unstable countries and the emission of pollutants.

Biofuels are nowadays the best alternative to substitute, or partially substitute petro-fuels. Bioethanol and biodiesel are recognized as the leading biofuels. A target of 10% biofuels usage has been fixed by the European Union by 2020.[4] It is clear that the necessity to optimize biofuels production processes is a priority to reduce the price difference with the petro-fuels. Considering only bioethanol, the first generation was produced from grains, contributing to the food vs fuel problem.[5] The production route has now been oriented to cheaper and non-food feedstocks like lignocellulosic biomass or sugarcane bagasse.[6,7] The typical bioethanol production process includes the steps: pretreatment, hydrolysis, concentration and detoxification, fermentation and finally product separation.[8]

Independent of the feedstock considered or the process technology used, it is always necessary to purify bioethanol from water to allow its use as a vehicle fuel. Bioethanol is miscible in all proportions with water and the presence of a homogeneous azeotrope makes the separation challenging.

Different separation processes have been proposed in the literature, from pressure swing,[9,10] passing through membrane separation[11] to biobased adsorbent[12] and diffusion distillation.[13] In addition, different hybrid technologies have also been proposed.[14−16] Among all, extractive distillation remains one of the most widely used alternatives for bioethanol dehydration due to its low energy consumption and large-scale production capacity.[17,18]

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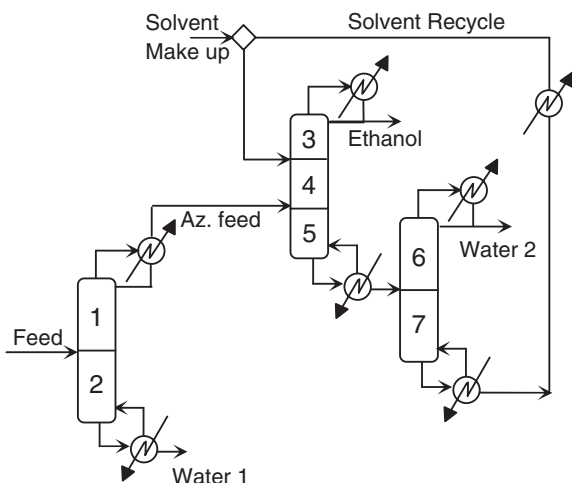
Figure 1. Classical extractive distillation sequence.

In extractive distillation a high boiling-point solvent is added as separating agent in order to increase the relative volatility of the components to be separated. Two main research areas are connected to extractive distillation: the selection of suitable solvents and the definition of new column configurations. In the former category computer aided molecular design (CAMD) represents a valid tool for the screening and selection of efficient solvents, reducing at the same time the experimental work.[19]

Regarding the last point, different configurations have been presented in the literature using simple columns,[20] complex columns,[21] or divided wall columns,[22] all aimed at the possibility of reducing the energy consumption and the capital investment. Due to the azeoreopic nature of the mixture and the multiple distillation columns used, researchers are still very actively looking for promising alternatives which are attractive in terms of either reduction of energy consumption or capital costs, or both.

Frequently the bioethanol purification problem is approached by testing different alternatives already reported in the literature and adapted to the specific azeotropic separation. Differently, in the present work, starting from analysis of the classical separation sequence composed of three columns, an alternative two-column sequence is introduced combining column sections performing analogous separation tasks. The proposed configuration is then studied for its steady state and dynamic performances.

The existing extractive distillation sequences

Extractive distillation is used to separate azeotropic mixtures by adding a solvent in the same column where the feed is introduced, usually called the extractive column. Another distillation column is necessary to recover the solvent that is recycled back to the extractive column. When the bioethanol process is considered, due to the dilution of the feed, the extractive column is preceded by a pre-concentration distillation column used to approach the azeotropic composition. The corresponding configuration is reported in Fig. 1, and is composed of three columns. It is possible to notice that this sequence has been developed following the heuristic rule that suggests removal of the mass separation agent in the separator immediately after the one into which it is introduced.[23] This sequence has been studied extensively in the literature for its optimal design.[24,25]

The possibilty to use a partial condenser in the pre-concentrator column in order to have a vapor feed in the extractive column was mentioned by Seader et al.[26] This alternative was reconsidered by Taylor and Wankat[27] together with recycling between the solvent recovery column distillate and the pre-concentration column.

More recently Li and Bai[28] proposed a configuration with a post-fractionator after the solvent recovery column. The principle used to develop this configuration derived from the equilibrium diagram for the ethanol–water system. The authors noticed that below 21% mol ethanol, the relative volatility of the system without the solvent is higher than the system with the solvent. This concentration value was set as the feed composition to the post-fractionator.

Starting from the Li and Bai's configuration Errico et al. developed a set of alternatives using different combinations of total and partial condensers,[20] thermally coupled sequences, and intensified sequences with a reduced number of columns.[21]

Kiss and coauthors extended the use of extractive distillation to different divided wall column arrangements obtaining promising results regarding the achievable energy savings.[22,29,30]

Synthesis of the alternative sequence

Distillation sequences synthesis is a procedure that allows the designer to move from the starting configuration (the reference) to all the alternatives predictable. Depending on the method or principle used, it is possible to identify different types of sequences mapping a more or less wide space of alternatives. In previous work[31] the thermal coupling technique and column section recombination was used as a method to generate a complete set of distillation alternatives starting from the subspace including all the simple column sequences. A thermal coupling is defined as bidirectional vapor and liquid streams that substitute a condenser or a reboiler associated with non-product streams located between two consecutive columns.[32] Examining the classical extractive distillation sequence of Fig. 1 it is possible to notice that following the forementioned method it is not possible to combine the pre-concentration and the solvent recovery column since they are not consecutive columns. The possibility of combination represents an interesting option since the same product is obtained from the stripping section of the pre-concentration column and from the rectifying section of the solvent recovery column. Referring to the column section notation of Fig. 1, theoretically it is possible to eliminate the reboiler and the condenser associated with section 2 and 6, respectively, merging the pre-concentrator and the solvent recovery column by means of a side stream. The overhead vapor from section 6 will substitute the vapor boil-up of the pre-concentrator's reboiler. At the same time the liquid from section 2 will substitute the reflux provided by the condenser of the solvent recovery column. The difference between the vapor overhead and the liquid flowrate corresponds to the water side stream flowrate. The corresponding configuration is reported in Fig. 2. This two-column configuration represents a novelty among all the configurations proposed for bioethanol purification discussed above. Compared with the classical extractive distillation column sequence reported in Fig. 1, the two-column configuration has one condenser and one reboiler less. Morover, only a single water stream is obtained.

Case study and steady state results

In order to prove the potential of the new configuration, a representative ethanol–water stream, hypothetically obtained from the fermentation step, is considered. Its physical properties and composition are reported in Table 1. The sequence was design
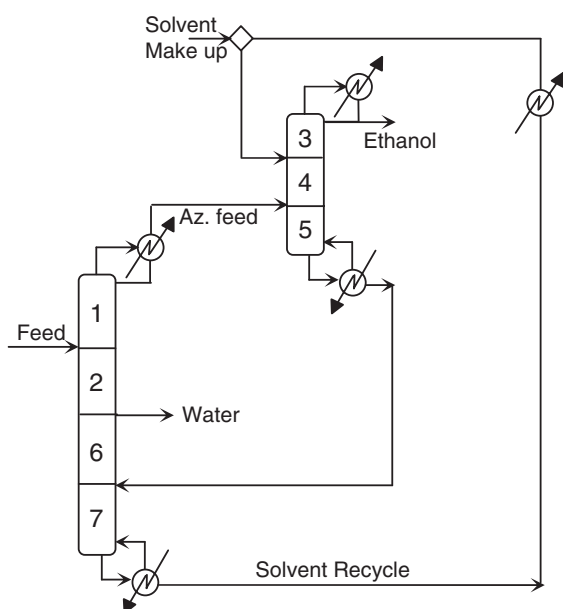
Figure 2. Two-column alternative sequence.

Table 1. Feed characterization

mole flow (kg h^{-1})	83870.7
pressure (kPa)	100
vapor fraction	0
enthalpy (GJ h^{-1})	−1209.034
composition (mass fraction)	
ethanol	0.121
water	0.879

Table 2. Design parameters, energy consumption and capital costs of the configuration in Fig. 1

	C_1	C_2	C_3
total number of stages	48	19	10
feed stage number	45	16	4
reflux ratio	5.65	0.80	0.71
solvent feed stage	---	3	---
column pressure (kPa)	100	100	100
column diameter (m)	3.10	1.47	0.67
distillate flowrate (kg h^{-1})	10662.10	9875.21	793.099
water 1 flowrate (kg h^{-1})	73208.60	---	---
water 2 flowrate (kg h^{-1})	---	---	793.099
solvent flowrate (kg h^{-1})	---	9095.79	---
purity bioethanol product (wt %)	---	99.9	---
purity of water 1 (wt %)	99.9	---	---
purity of water 2 (wt %)	---	72.0	---
purity of ethylene glycol recycle (wt %)	---	---	1.00
condenser duty (kW)	−15474.50	−4185.35	−724.64
reboiler duty (kW)	19103.00	2152.62	1170.68
total condenser duty (kW)	−20384.41		
total reboiler duty (kW)	22426.30		
annualized capital cost (k\$ y^{-1})	198.8		

Table 3. Design parameters, energy consumption and capital costs of the configuration in Fig. 2

	C_1	C_2
total number of stages	58	19
feed stage number	45	16
reflux ratio	5.64	0.80
side stream stage	52	---
solvent feed stage	---	3
column pressure (kPa)	100	100
column diameter (m)	3.15	1.48
distillate flowrate (kg h^{-1})	10782.50	10050.10
water flowrate (kg h^{-1})	73845.30	---
solvent flowrate (kg h^{-1})	---	9095.51
purity bioethanol product (wt %)		99.90
purity of water (wt %)	99.8	---
purity of ethylene glycol recycle (wt %)	1.00	---
condenser duty (kW)	−15495.40	−4259.45
reboiler duty (kW)	19572.70	2226.97
total condenser duty (kW)	−19754.85	
total reboiler duty (kW)	21799.67	
annualized capital cost (k\$/y)	178.9	

to obtain 99.9% wt pure ethanol in order to satisfy the requirements for its use as vehicle fuel.[33] The minimum purity of water was defined as 99.8% wt in order to optimize its use inside the plant.[34]

The NRTL method was chosen to evaluate the non-ideality of the liquid phase. Ethylene glycol was selected as entrainer and its make-up flowrate was minimized in order to preserve the economics of production. All calculations were performed using the process simulation package Aspen Plus V8.0. The initial design parameters for all the columns were obtained from the work of Errico and Rong[35] and then optimized to achieve the product purity requirements. The capital costs were evaluated with the Aspen Economic Evaluator package, recognized as the most reliable tool for the estimation of equipment cost.[36] Sieve trays columns, fixed tube condenser and kettle reboilers are considered for the evaluation. The total condenser and reboiler duties are used as surrogates to the energy cost.[32] The results for the classical configuration of Fig. 1 are reported in Table 2. The design of the two-column sequence, shown in Fig. 2, was defined according to the correspondence in the column sections´ function among the classical separation sequence of Fig. 1. This means that the first column of Fig. 2 is obtained by combining the first and the third colum of Fig. 1.[37] The side stream water flowrate is in liquid phase and its flowrate was defined according to the water content of the feed and the purity specification for the solvent stream recovered from the bottom. Optimum feed and side stream locations were checked using the sensitivity analysis implemented in the Aspen Plus simulator. The corresponding design, together with the

energy consumption and the capital cost evaluation, is reported in Table 3. Notice that the two-column sequence has an energy consumption 3% less than the traditional configuration, more significantly a 10% saving in capital cost is achieved. Regarding the productivity, the two-column sequence realizes 99% ethanol recovery compared with the 97% for the classical configuration.

Control properties

One of the key considerations for the dynamic analysis is the selection of control outputs and manipulated variables for each control loop. Although more formal techniques to define the
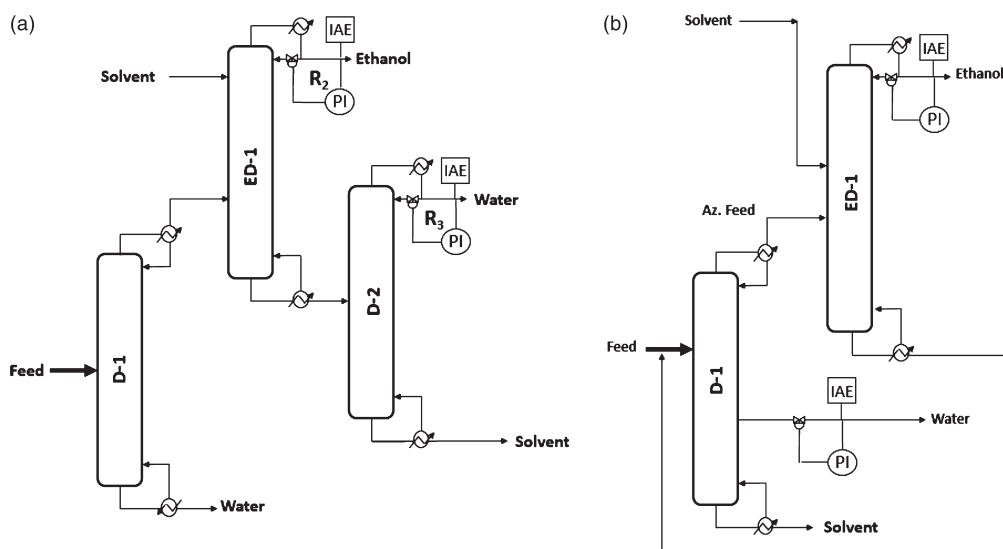
Figure 3. Loops LV: (a) classical separation sequence (BC); (b) two-column sequence (MTC).

control loops for the integrated columns may be used, the selection was made based on practical considerations. A well known structure is based on energy balance considerations, which yields to the so-called LV control structure in which the reflux flowrate L and the vapor boilup rate V (affected directly by the heat duty supplied to the reboiler) are used to control the distillate and bottom output compositions.[38] It should be mentioned that such control loops have been used with satisfactory results in previous studies on complex distillation systems.[39−41] Thus, for any sequence, the control of the lightest component (ethanol) was manipulated with the top reflux flowrate, and the water in the classical configuration with the top reflux flowrate whereas the water in the two-column sequence was controlled with side stream flowrate as reported in Fig. 3. The closed loop analysis was based on proportional-integral controllers. Several alternatives exist for tuning the controller parameters. We attempted a common ground for comparison by optimizing the controller parameters, proportional gains (K_C) and reset times (τ_i), for both the traditional and the two-column schemes following the integral of the absolute error (IAE) criterion. For the two-column arrangement, the procedure is particularly complicated because of the interactions of the multivariable control problem. In this case, the tuning procedure was conducted taking one control loop at a time; the parameters thus obtained were taken for the following control loop until the two loops were considered. For the dynamic analysis, individual set point changes for product composition were implemented for each of the product streams. For all cases (classical and two-column sequence), the two control loops were assumed to operate in a closed loop fashion.

Dynamic results

The performance of the sequences under analysis was compared through the evaluation of IAE values for each test. This part of the study was conducted with the use of Aspen Dynamics.

Table 4 shows the IAE values obtained for each composition control loop of the two distillation sequences. The sequence with two columns offered the best dynamic behavior, based on the lowest values of IAE, for control of the two product streams. The individual dynamic responses of each control loop for the two distillation sequences are displayed in Fig. 4. As observed from the figure, the control of ethanol or water does not create any

Table 4. Optimum closed loops LV

	Component	K_C	τ_i	IAE
BC	Ethanol	250	2.50	0.001075393
	Water	1.00	78.00	0.092859003
MTC	Ethanol	250	2.50	0.001032373
	Water	150	1.00	0.000084499

significant problem for any of the two sequences, although the configuration with two columns showed the lowest IAE values.

CONCLUSIONS

A two-column sequence was proposed as a new alternative to the traditional three column configuration for the separation of bioethanol by extractive distillation. This new sequence was obtained considering the similarities in the column sections performing the same separation task.

It is interesting to notice that for the case study considered, the two-column configuration offers the lowest capital cost, the best dynamic performance and almost the same energy consumption compared with the traditional sequence. The main results obtained for the case considered are summarized in Table 5. The control performance and the energy consumption are compared together with the carbon-dioxide emission calculated as proposed by Gadalla *et al.*[42] This analysis was included because sometimes it is possible to obtain the best dynamic performance operating with high CO_2 emission levels. The specific energy requirements, defined as the energy used per kg of bioethanol produced, is also reported. The two-column sequence has a 4.5% saving in the energy requirement. The two-column sequence proposed represents a new valid alternative for the separation of bioethanol by extractive distillation. It is able to operate with one condenser, one reboiler and one column less than the classical sequences, reaching 10% reduction of capital cost with a sligtly lower energy consumption but with higher ethanol recovery. The dynamic analysis also confirms that the two-column sequence is superior to the classical configuration.
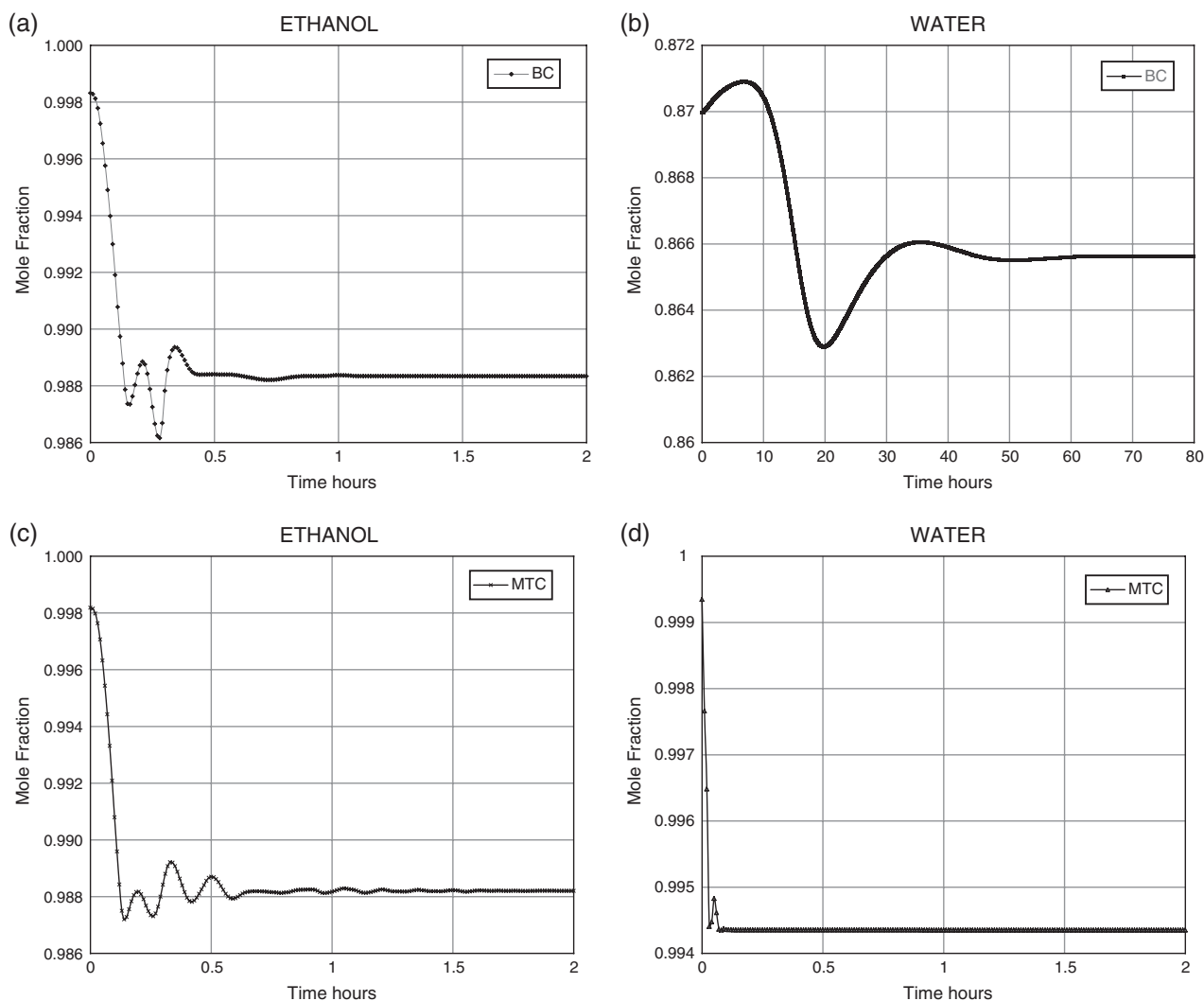
Figure 4. Dynamic responses: (a, b) classical separation sequence (BC); (c, d) two-column sequence (MTC).

Table 5. Optimum closed loops, energy consumption, CO_2 emissions and specific energy usage

	Control		Total Reboiler duty	CO_2 Emissions	Specific energy requirements
	IAE Ethanol	IAE Water	(kW)	(ton h^{-1})	(kW h kg^{-1})
BC	0.001075393	0.092859003	22426.30	126.890	2.271
MTC	0.001032373	0.000084499	21799.67	123.030	2.169

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