



Energy saving alternatives for renewable ethanol production with the focus on separation/purification units: A techno-economic analysis



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ABSTRACT

The bioethanol production process includes energy-demanding units with high operating costs, i.e., separation, dehydration, and stillage evaporation units. The present study attempts to reduce energy consumption and, ultimately, ethanol price through industrially feasible methods. These methods include adopting thermally-coupled columns, dividing-wall columns, single/multi-stage pervaporation process, and evaporators. Nine scenarios were studied and compared with an industrial process case using heat-integration and thermal vapor recompression system. Each scenario was simulated with Aspen Plus® and analyzed with Aspen Process Economic Analyzer. The pervaporation process with a hydrophilic polymer membrane was modeled with MATLAB using sorption-diffusion theory and three-conservation law. Then, the required membrane area and the temperature drop during the pervaporation process were estimated. The dividing-wall column reduced the separation unit's energy consumption by 67% and the capital costs by 19%. The energy consumption decreased by 49% by adopting thermally-coupled and heat-integrated columns, and capital costs were reduced by 17%. Moreover, the four-stage pervaporation process for ethanol dehydration proved to be more profitable than the pressure swing adsorption. The proposed and investigated methods reduce the cost associated with the processes' energy consumption and decrease bioethanol's overall production cost.

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1. Introduction

With increasing greenhouse gases (GHG) emission, oil price fluctuations, and increasing demand for energy, particularly in the transportation sector, scientists and manufacturers have been thinking of using alternative renewable energy sources [1]. Approximately 85% of the world's energy demand is provided by non-renewable and fossil fuels, leading to serious environmental problems such as ozone depletion and climate change [2,3]. These problems necessitate using renewable fuels as a sustainable energy resource for long-term use.

Renewable fuels are regarded as solar, wind, hydrothermal, geothermal, and biofuels. Liquid biofuels are promising alternatives for gasoline and diesel [3,4]. Among different types of biofuels, bioethanol can be blended with gasoline due to its high octane and

low cetane numbers, which thus results in better oxidation of hydrocarbons. Thus, bioethanol is a renewable and eco-friendly substitutive fuel to gasoline [5,6].

Industrial bioethanol is produced from biomass, either in the form of free sugars, starch, or lignocellulose. Biomass in the form of free sugars, such as sugarcane, is globally used to produce ethanol at large scales due to the lowest total production cost and fossil energy consumption [5]. The production process of bioethanol from sugarcane includes five main units: feed handling, fermentation, separation, dehydration, and stillage evaporation. Separation, dehydration, and stillage evaporation processes of bioethanol production consume more than 88% of the total required energy and takes 5.6% of the total cost for the first- and second-generation ethanol based on provided industrial data by Sepahan Biological Company, Isfahan, Iran, and the report by Humbird et al. [6]. This report was prepared by National Renewable Energy Laboratory (NREL) and is the most widely benchmarked reference regarding ethanol production from lignocellulosic biomass. There are uncertainties about the high energy consumption and, consequently,

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operating costs of separation, dehydration, and stillage evaporation processes of bioethanol production. This high energy consumption could be one reason why there are still questions about bioethanol's renewability [7–9].

Several studies presented energy and cost reduction for the bioethanol production process; however, the research areas have been predominantly restricted to the dehydration unit. Brito et al. [10], Tututi-Avila et al. [11], Erico and Rong [12], Kiss et al. [13], and Tavan et al. [14] utilized extractive and azeotropic distillation to overcome the separation problems of the water and ethanol azeotropic mixture (95.6% w/w ethanol at 78°C). However, because of the complexity, excessive energy loss, and high expenses, the application of traditional azeotropic and extractive distillation is limited in the industry [15]. Consequently, manufacturers have been questing for an efficient alternative method for bioethanol dewatering, such as pressure swing adsorption (PSA) with molecular sieve and multi-stage pervaporation [16]. Compared with extractive and azeotropic distillation, a pervaporation module results in better selectivity, lower energy consumption, and more profitability. Furthermore, pervaporation is known as an environment-friendly dehydration technology. This method neither needs any additives, e.g., entrainer additive in extractive and azeotropic distillations, nor any adsorbent such as PSA [17].

Besides dehydration, separation and evaporation units can play a vital role in ethanol plants' energy optimization. The separation process can be developed through column heat integration and different column sequences, namely, thermally-coupled columns, dividing-wall columns, and hybrid pervaporation/distillation. The heat integration concept is based on the exchange of heat from hot streams with higher enthalpy to cold streams with lower enthalpy. In this study, column heat integration was the heat integration between the reboilers and condensers of columns and not other streams. As a dehydration method, the pervaporation process is not restricted by thermodynamical constraints, e.g., azeotrope of water and ethanol. Thus, it results in lower energy demand compared with the distillation column in the separation unit [17]. Moreover, the fossil energy ratio (FER) of the stillage evaporation unit is improved through the application of mechanical vapor recompression (MVR) or thermal vapor recompression (TVR). Sugar beet molasses is the most dominant feed for ethanol production, and natural gas is the most available and used fuel for energy production in ethanol plants in Iran [18]. In applying a TVR or MVR system, operational costs of steam or electricity should be considered [19]. Analysis for energy reduction in ethanol production processes was performed by Lopez-Castrillon et al. [20], Pina et al. [21], and Cortes-Rodriguez et al. [22]. However, previous studies in this field do not appear to be concerned about applying new industrial methods. There is a high potential for the improvement of FER and profitability index of the bioethanol production process in this regard [2].

The present research sets out to propose a conceptual design of a novel bioethanol production process focusing on reducing energy consumption and costs. To this end, nine scenarios in separation, dehydration, and evaporation units are proposed and contrasted with one another. For bioethanol separation, a three-column Othmer distillation system, which is the most commercial distillation unit for ethanol separation worldwide, is chosen along with different column operating pressures [23]. Due to the difference in operating pressures, heat loads of condensers and reboilers are integrated efficiently. Furthermore, thermal coupling and dividing-wall are applied in a multi-column system for better thermal and economic efficiency. The behavior of pervaporation process for solvent dehydration was successfully described with the sorption-diffusion mass-transfer mechanism [24]. The pervaporation module was modeled by MATLAB based on the three-conservation law

[25] with the application of the sorption-diffusion mass-transfer mechanism. The applicability of the model is proved and tested with laboratory-scale experiments for water-ethanol mixtures. For the water-ethanol separation using a hydrophilic polymeric membrane, the required membrane area and the temperature drop during the pervaporation module are estimated with the developed model [25]. Each scenario was simulated and analyzed economically with Aspen Plus and Aspen Economic Analyzer software, respectively, using industrial data from a commercial plant. A sensitivity analysis was then conducted to assess the most affecting parameters and the influence of the proposed scenarios on the total production cost of bioethanol.

2. Method

The commercial base case process and nine alternative scenarios were simulated with Aspen Plus® considering the defaults of Aspen Plus, a convergence of 0.0001, and a maximum iterations number of 30. This software is known as one of the most useful commercial programs for the simulation of processes. This software simulates processes through a sequential-modular program with different mathematical modeling such as the Rstoic model for reactors as well as the Radfrac model for distillation columns. Distillation columns have a degree of freedom of two once the feed, the feed tray location, and operating pressure have been specified. Through a Radfrac model, an equilibrium-stage approach using mass, equilibrium, and summation of heat equations was used. For ideal systems, it is possible to estimate the number of stages, reflux ratio, and feed stage using short cut methods, i.e., the Winn-Underwood-Gilliland method. However, this method is not recommended for non-ideal water-ethanol mixtures. Thus, the initial data of distillation column was adopted from a commercial ethanol plant, Sepahan Biological Products Company, Isfahan, Iran. After having the column converged, the optimization of the parameter was conveyed to have a product recovery of 99% in each column. Thus, there is some ethanol loss from column bottoms. However, the cost for recovery of this 1% ethanol is more than its value. This cost involves capital investment to have a larger column and energy costs. The number of stages is an indicator of column capital cost, and the reflux ratio is an indicator of operating costs (utility costs). Therefore, a near-optimum point for reflux ratio and the number of trays was investigated to minimize the total column cost.

In this study, the vapor-liquid-equilibrium (VLE) data were predicted with high precision through the use of non-random two-liquid (NRTL) as the base method [26]. NRTL model with three parameters is an activity coefficient model that estimates liquid phase fugacity of binary and multi-component mixtures. For non-ideal gases, the Soave-Redlich-Kwong (SRK) equation of state (EOS) was used. Besides, the properties of non-condensable gasses, e.g., carbon dioxide and methane, were calculated by Henry's Law [27,28]. The rates and prices for raw materials and products are presented in Table S1. Furthermore, the unknown binary interactions were estimated by the UNIFAC group-contribution method [29]. Bio-components, e.g., glucan, xylan, mannan, galactan, ash, and lignin, were manually introduced to the Aspen Plus databank based on the report by Wooley and Putsche [26].

The sizes of equipment items were estimated using mass and energy flow rates from the simulation. Furthermore, the required utilities were calculated based on the simulation results. The equipment price data bank of Aspen PEA and local manufacturer quotes was used to estimate the equipment costs [30]. Then the total capital investment was calculated based on the equipment's direct cost and by Aspen PEA. The plant operating costs were calculated using Aspen PEA and the required utilities. The scenarios' economic assessment was performed with Aspen Process

Economic Analyzer (Aspen PEA) using the mass and energy results from Aspen Plus and total capital investment and operating costs. The most important assumptions are presented in Table S2. Due to the underestimation of Aspen PEA for individual equipment, such as the evaporator, the cost of such an item was obtained through exponential scaling expression based on the cost data given in Davis et al. [31], instead of Aspen PEA. The scaled equipment items' costs were indexed to the 2016 USD using the chemical engineering plant cost index. The utility costs, namely electricity, cooling water, and steam at over 7 atm, were provided by Fajr Petrochemical Company, Imam Khomeini Port, Iran [32], and local manufacturers. The electricity is bought from the grid. While steam is locally generated using natural gas and diesel.

Considering the obtained economic features from Aspen PEA, a sensitivity analysis was conveyed to investigate the most affecting parameters and the influence of nine scenarios and the base case on the bioethanol total production cost.

2.1. Base case description

Data for the base case was adopted from a commercial ethanol plant, Sepahan Biological Products Company, Isfahan, Iran. In this plant, bioethanol is produced at a rate of 227 kg/h from 1130 kg/h of sugarcane molasses composed of 51%(w/w)water, 47%(w/w) sucrose, and 2%(w/w) glucose coming as a byproduct from the sugar industry. Based on the commercial plant, the ethanol production process from sugarcane molasses involves six basic units: feed handling, fermentation, separation, dehydration, stillage evaporation, and storage units (Fig. 1).

In the feed handling unit, the raw material is mixed with water in the ratio of 2:3 to reach 18% (w/w) sugar concentration, including sucrose and glucose. This concentration is lower than the maximum sugar content, which inhibits the fermentation, i.e., 20% (w/w) [33].

In the batch fermentation unit, sugar, mainly sucrose, is converted to monosaccharides consumed by the yeast *Saccharomyces cerevisiae*. The fermentation temperature and residence time are set to be 30 °C and 45 h. In addition, the fermentation reaction yield of ethanol production is assumed to be 90% [34,35]. The fermentation is operated in batch mode of operation; however, this unit is scheduled to facilitate a continuous product stream for the distillation unit. The fermentation was simulated as a continuous reactor using the Aspen Plus® Rstoic model, considering reaction stoichiometries and conversion factors. The total volume of fermenters was calculated assuming 80% working volume and a residence time of 45 h plus filling and emptying time [34]. With a total volume of

172 m³, the three fermentation reactors are made of clad carbon to prevent the effect of corrosive materials in this unit [36]. Organic acids, aldehydes, and 9.5% (w/w) ethanol are the fermentation unit products. A stream containing carbon dioxide, saturated with water and ethanol vapors, is separated from the top of fermenters.

Meanwhile, organic acids, aldehydes, and 9.5% (w/w) ethanol are obtained from the fermenters' bottom stream. The fermentation broth is pumped to the distillation unit continuously by scheduling the batch fermentation. The base case's distillation unit involves a three-column Othmer system, which was simulated with the Radfrac distillation model in Aspen Plus. This model handles rigorous stage-by-stage mass and energy balance calculations based on phase equilibrium data. The column sequences were chosen based on design heuristics for simple columns. In this design, water with the largest fraction of distillation feed was first separated. Then, the azeotropic mixture, 95.6 wt % ethanol at 78, which has the most difficult separation, is removed in the last column [29,37]. The first column purifies ethanol to 57.2% (w/w). The second and third columns are responsible for the removal of light components, and production of ethanol with the purity of 94% (w/w), respectively. Then, the overhead of the third column is pumped to the dehydration unit for further purification.

For the dehydration of ethanol, a pressure swing adsorption system with a molecular sieve is implemented. The molecular sieve consists mainly of synthetic zeolite with 3°A molecular pores, while water and ethanol molecular pores are 2.8° A and 4.2°A, respectively. The schematic of this process is presented in Fig. 2A.

The bottom product of the first distillation column, containing water, organic acids, and aldehydes, is sent to the stillage evaporation unit. In this unit, the stillage is concentrated to vinasse, i.e., a thick liquid containing 60% dry solid content, e.g., ash, and 40% liquid, including mainly water and biomass. For this purpose, a falling film evaporator followed by a forced circulation evaporator was used (Fig. 2A).

2.2. Scenarios description

Separation, dehydration, and stillage evaporation units are among the most energy consumers and costly units of commercial bioethanol production plants. Based on industrial data, more than 88% of the consumed energy in the bioethanol plants is spent in the mentioned units. For the purpose of optimization of the base case, nine scenarios for separation, dehydration, and stillage evaporation units were proposed. For these scenarios, the energy consumption and economic profitability parameters, namely, internal rate of return, profitability index (PI), capital costs, payout period, and

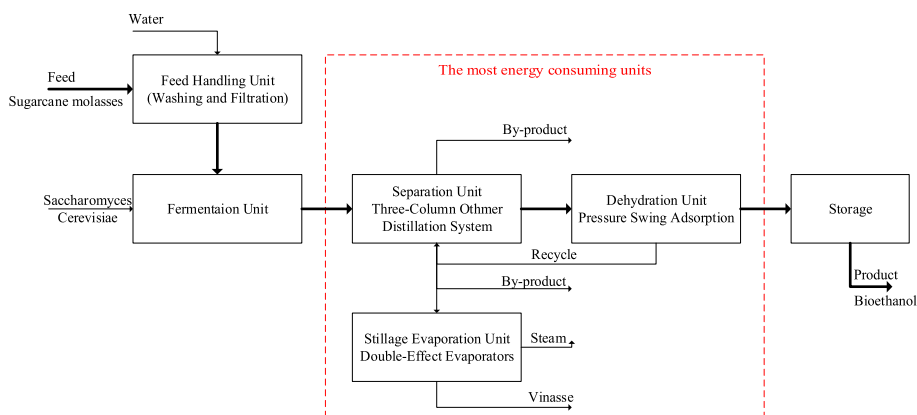


Fig. 1. Main process units of the bioethanol production process from sugarcane molasses.

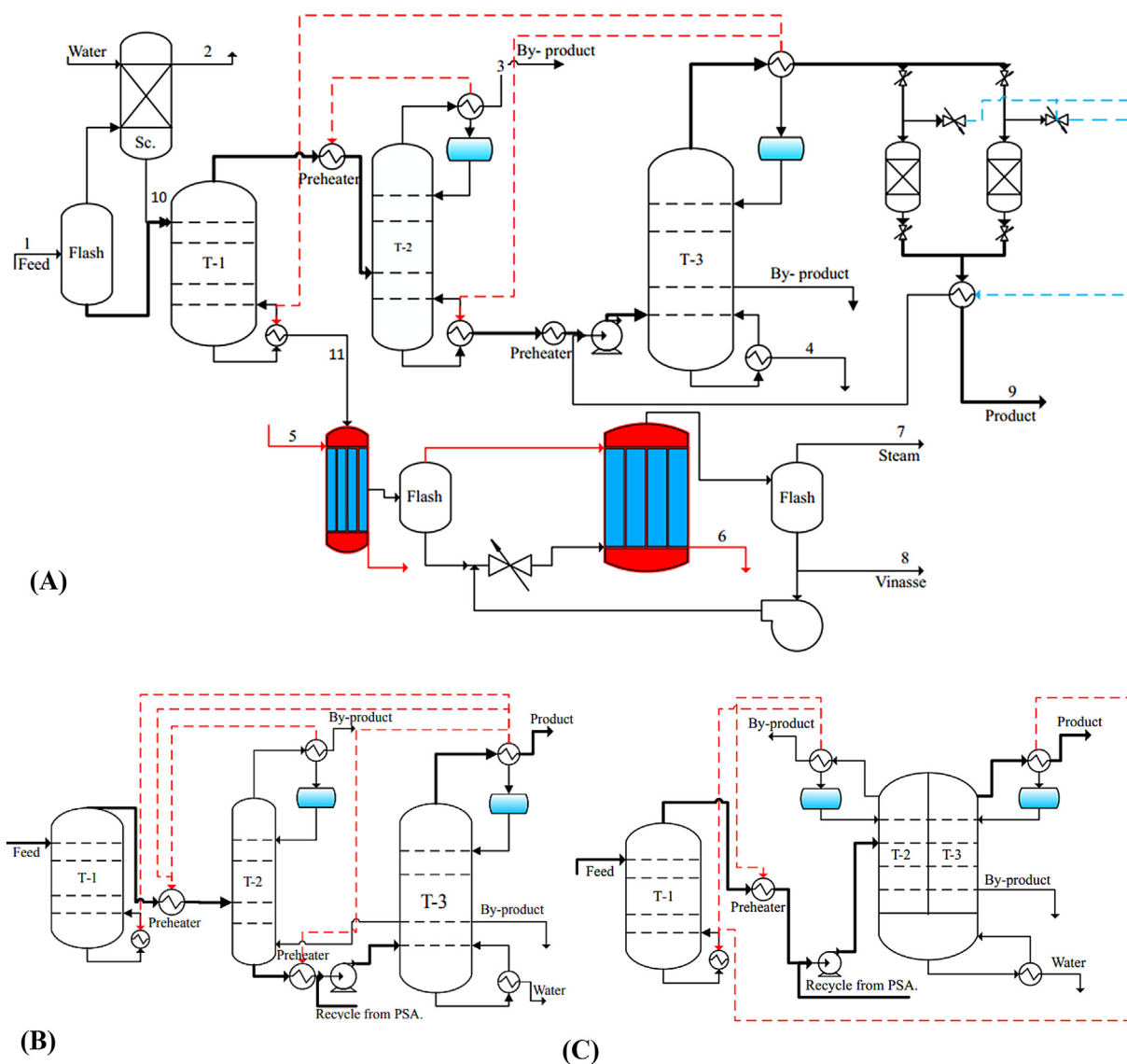


Fig. 2. (A) Scenario 1, application of multi-pressure distillation columns with heat integration, molecular sieve unit for ethanol dehydration, and two-stage evaporation system for the first column stillage, (B) Scenario 2, application of thermally-coupled distillation with heat integration, and (C) Scenario 3, distillation columns by using dividing-wall column for the second tower with heat integration.

total production cost of ethanol, were calculated and compared for all scenarios. All heat exchangers in the distillation unit were assumed to be shell and tube heat exchangers for performing heat integration on the same devices. Differences in scenarios are in the separation, dehydration, and stillage evaporation units, which are explained as follows:

2.2.1. Scenario 1

In scenario 1, the base case with multi-pressure columns is integrated thermally (Fig. 2A). In this scenario, thermal energy is recovered from cooling and heating streams in distillation columns with different operating pressures, which play a key role in the column heat integration. The heat integration concept is based on the exchange of heat from hot streams with higher enthalpy to cold streams with lower enthalpy so that the energy sources of the process are used efficiently [38]. As the red dashed lines show in Fig. 2A, the reboiler duty of T-2 with an operating pressure of 0.8 atm is provided by the released latent heat from condensation of the top product in the distillation column T-3 with an operating

pressure of 1.8 atm. The rest of obtained energy from T-3 is used in the reboiler of T-1 with an operating pressure of 0.3 atm.

2.2.2. Scenario 2

In scenario 2, the second and third distillation columns (rectifier) are coupled thermally in the integrated three-column Othmer distillation system (Fig. 2B). In this scenario, the required heat for reboiling in the distillation column T-2, with an operating pressure of 0.8 atm, is supplied by the partial condensation of the top vapor stream from the high-pressure column T-3. This heat is transferred in a shell and tube heat exchanger. Therefore, the reboiler of the T-2 column would be one of the condensers for the T-3 column. Thus, the capital and operating costs of a reboiler in column T-2 are saved compared with the conventional distillation system.

2.2.3. Scenario 3

In scenario 3, the dividing-wall column is used as an alternative for an energy-intensive conventional distillation column sequence (Fig. 2C). In this scenario, the desired components are separated in a

single column instead of two columns since two columns need higher capital and operating costs. Thus, the second and third columns, T-2 and T-3, are converted to one column through the partition wall along with column heat integration (Fig. 2C). Heat transfer over the dividing-wall can lower the column performance. Thus, the dividing-wall is thermally isolated. Since the cost of the dividing-wall column is not provided by Aspen PEA, one column is assumed as the dividing-wall column with the equivalent tray number and area for the cost estimation. Despite this assumption, all the required equipment and parts are considered in the economic evaluation through the calculation of two circle segments to have a minimum error (see Table 3).

2.2.4. Scenario 4

In scenario 4, pervaporation process uses a commercial hydrophilic membrane, PERVAP2210, supplied by Sulzer Chemtech. As Fig. 3 demonstrates P1-4, the membrane separation process is used as a substitute for PSA to dehydrate the azeotropic water/ethanol mixture and produce ethanol with a purity of 99.5% (w/w). Pervaporation process is modeled and optimized through a developed process design procedure in the MATLAB environment (version 2015b). The selected design algorithm sets out to propose a steady-state flow model based on the three conservation law mass and energy with the following assumptions:

- The sorption-diffusion mechanism, which is widely accepted for introducing the mass transfer mechanism through non-porous membranes. This mechanism includes three steps of sorption on the feed side, diffusion through the membrane, and desorption on the product side [39]. A published experimental data set, namely the enrichment coefficient, water content in the feed, and permeate flux at different temperatures, was used in order to estimate the parameters of the flux model [40]. After the estimation of the model parameters, the model was implemented for our case.
- The feed at the liquid phase is thermostated at the required temperature before entering the pervaporation module.

The details of this model were presented by Vatankhah [25].

2.2.5. Scenarios 5-9

Scenarios 5–9 are applied to assess the influence of the number

of evaporation effects and TVR system on the economy and required fresh steam of the stillage evaporation unit. To achieve a better evaporation economy, the TVR system, an inexpensive technology, is implemented due to the availability of the high-pressure steam over 7 atm, which creates a partial vacuum by pumping vapor from the evaporator. The resultant pressure is lower than the motive steam and higher than the vapor from the evaporator. The compression ratio was calculated using expansion ratio and the kilogram of motive steam per kilogram of load [41]. The compression ratio is the ratio of the discharge pressure to the suction pressure, while the ratio of the motive pressure to the suction pressure was considered as the expansion ratio. The operational costs for this system are then compared with the base case and other scenarios. In scenario 5, a single-effect evaporator is used in place of double-effect evaporators in the base case, while in scenario 6, a single-effect evaporator with a TVR system is applied (Fig. 4). In scenario 7, the TVR system is used in conjunction with double-effect evaporators. In scenario 8, triple-effect evaporators are utilized to investigate the impact of the number of evaporators on the evaporation unit's operating and capital costs. The more the evaporation effects, the better steam economy is obtained, and the higher capital cost is required. In scenario 9, the impact of TVR system on the triple-effect evaporators is studied.

3. Results and discussions

Due to the excessive energy consumption and operating costs of separation, dehydration, and stillage evaporation units in the base case, nine scenarios were studied and discussed to optimize the bioethanol production cost. In the separation and dehydration units, four scenarios, namely, integrated multi-pressure columns (scenario 1), thermally-coupled columns (scenario 2), dividing-wall column (scenario 3), multi-stage pervaporation (scenario 4) were investigated along with column heat integration. The column pressures were selected to guarantee the use of cooling water in the condensers. The condenser temperature for all columns is more than 60 °C (Tables 1 and 3) which allows at least 30 °C temperature difference with the cooling water. In addition to the separation and dehydration units, five scenarios, single-effect and multi-effect evaporators without and with TVR system, were surveyed in the stillage evaporation unit.

Table 1

Specifications of distillation columns for scenario 1^e (cf. Fig. 2A).

Columns	Scrubber	T-1	T-2	T-3
Top product, %(w/w) ethanol	minor	57.2	75.0	94.0
Bottom product, %(w/w) ethanol	2.3	0.1	55.0	0.3
Tray-type	random packings-Rasching rings	sieve	sieve	sieve
Feed stage	1	1	29	80
Number of stages ^a	5	29	49	88
Column diameter (m)	0.3	0.8	0.3	0.6
Ethanol recovery (%)	99.0	99.2	86.6	98.7
Condenser operating pressure (atm)	1.0 ^b	0.3 ^b	0.8	1.8
Condenser operating temperature (K)	297.5 ^c	335.2 ^c	345.2	366.5
Reflux ratio	—	—	0.2	6
Condenser duty without heat integration (kW)	—	—	−3.3	−374.7
Reboiler duty without heat integration (kW)	—	345.5	22.9	454.1
Total duty without heat integration ^d (kW)	1200.5			
Total cooling duty with heat integration ^d (kW)	−28.4			
Total heating duty with heat integration ^d (kW)	454.1			

^a A 60% tray efficiency was considered in the Aspen Plus simulation.

^b Pressure of the top stage.

^c Temperature of the top stage.

^d Total duty takes into account hot and cold utilities.

^e The initial data for trial-and-error calculation of distillation columns was adopted from a commercial ethanol plant, Sepahan Biological Products Company, Isfahan, Iran, for having a reliable accuracy.

Table 2

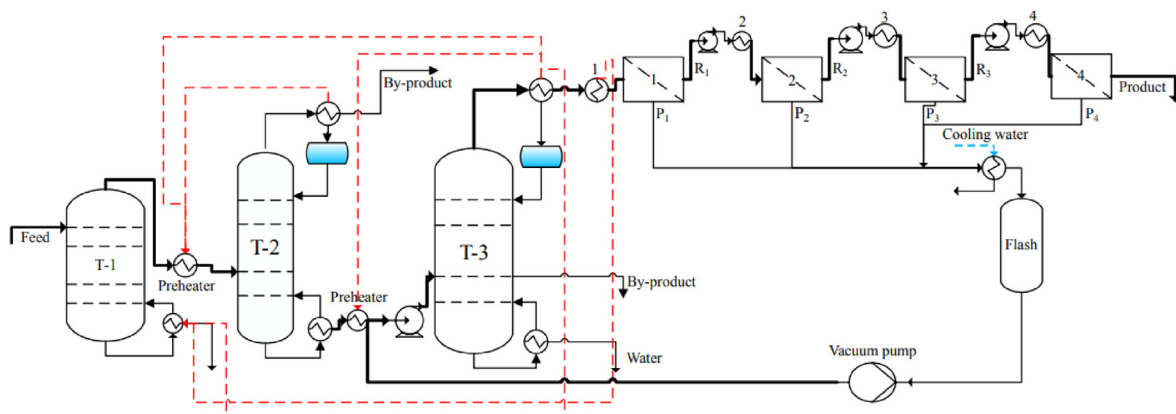
Stream specifications of the scenario 1 for separation, dehydration, and stillage evaporation units (cf. Fig. 2A).

Stream component	1	2	3	4	5	6	7	8	9	10	11
Major components (wt%)											
Ethanol	9.0	minor	75.0	0.3	—	0.2	minor	minor	99.5	9.0	minor
Water	76.3	1.0	14.0	99.6	100.0	99.8	99.9	9.5	0.5	85.0	92.0
CO ₂	8.0	99.0	9.0	—	—	—	—	—	—	0.1	minor
Biomass	2.0	—	—	—	—	—	—	25.0	—	2.0	2.1
Ash ^a	4.0	—	—	—	—	—	—	57.5	—	4.0	5.0
Minor components (wt%)											
Acetic acid	0.001	—	—	—	—	0.001	0.002	—	—	0.600	0.002
Glycerol	0.600	—	—	—	—	—	0.002	8.000	—	—	0.700
Methanol	0.005	—	0.040	—	—	0.003	—	—	—	0.004	0.001
1-Propanol	0.003	—	0.020	0.008	—	—	—	—	—	0.003	—
Isopropyl-alcohol	0.003	—	0.020	—	—	—	—	—	—	0.003	—
N-Butanol	0.003	—	0.006	0.010	—	—	—	—	—	0.003	—
3-Methyl-1-Butanol	0.010	0.002	0.020	0.100	—	—	—	—	—	0.010	—
2-Pentanol	0.002	—	—	0.010	—	0.004	—	—	—	0.002	0.002
Acetaldehyde	0.020	0.030	0.900	—	—	—	—	—	—	0.010	—
Formaldehyde	0.008	0.010	0.500	—	—	—	—	—	—	0.007	—
Total flow (kg/hr.)	3142.5	254.8	47.4	184.4	1335.0	1189.6	1206.2	221.1	227.8	3081	2617.0
Pressure (atm)	1.0	1.0	0.8	2.0	9.9	0.2	0.1	0.1	1.0	1.0	0.4
Temperature (K)	303.1	297.5	345.2	393.0	453.1	338.2	322.6	322.6	303.1	303.1	350.4

^a Ash contains mostly calcium-oxide.**Table 3**

Product stream specification and separation unit thermal results the for scenarios 2–4.

Scenarios	Scenario 2			Scenario 3			Scenario 4		
Columns	T-1	T-2	T-3	T-1	T-2	T-3	T-1	T-2	T-3
Top product, %(w/w) eth.	57.2	73.0	94.0	57.2	83.0	93.0	57.2	75.0	90.8
Bottom product, %(w/w) eth.	0.1	53.0	minor	0.1	minor	—	0.1	55.0	2.4
Ethanol recovery (%)	99.2	87.5	95.0	99.2	88.4	77.0	99.2	86.6	98.1
Tray numbers ^a	29	50	88	29	59	80	29	49	65
Column diameter (m)	0.8	0.3	0.6	0.8	0.3 ^b	0.45 ^b	0.8	0.3	0.4
Condenser pressure (atm)	0.3 ^c	0.8	1.8	0.3 ^c	1.8	1.8	0.3 ^c	0.8	1.8
Reflux ratio	0	0.2	6	0	1	4	0	0.2	6
Condenser temperature (K)	335.2 ^d	345.8	366.5	335.2 ^d	366.5	366.5	335.2 ^d	345.2	366.7
Reboiler temperature (K)	350.4	352.1 ^e	393.8	350.4	373.9	—	350.4	351.9	390.2
Condenser duty without CHI ^f (kW)	—	−1.5	−468.5	—	−25.5	−127.7	—	−3.3	−150.9
Reboiler duty without CHI ^f (kW)	345.5	—	502.9	345.5	195.5	—	345.5	22.9	163.9
Total duty without CHI ^f (kW)	use	—	—	694.2	—	—	686.5	—	—
Total cooling duty with CHI ^f (kW)	106	—	—	0	—	—	0	—	—
Total heating duty with CHI ^f (kW)	502.9	—	—	396.2	—	—	404.7	—	—

^a The tray numbers were calculated considering a tray efficiency of 60%.^b Columns T-2 and T-3 are circle segments instead of being a complete circle. The cross-sectional areas for these segments were 0.07 and 0.16 m². The stated diameters, 0.3 and 0.45 m are the equivalent diameter of a complete circle equivalent to the cross-sectional area of 0.07 and 0.16 m², respectively.^c Pressure of the top stage.^d Temperature of the top stage.^e Temperature of the bottom stage.^f CHI: Column heat integration.**Fig. 3.** Separation and dehydration unit of scenario 4 using a four-stage pervaporation module in the dehydration unit with column heat integration based on the heat exchange of hot and cold streams between the reboilers and condensers of columns.

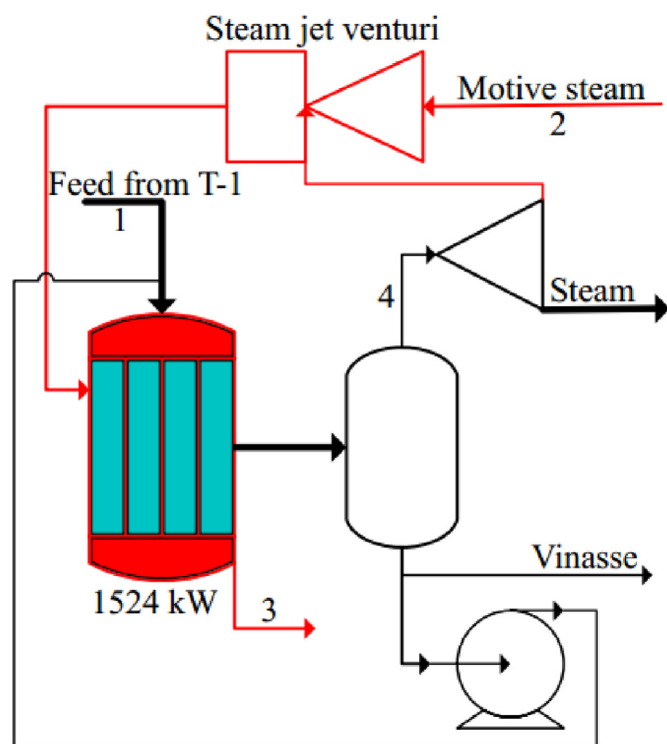


Fig. 4. Single-effect evaporator with TVR system for scenario 6.

3.1. Scenario 1

The conceptual design of integrated separation, dehydration, and stillage evaporation units of scenario 1 is shown in Fig. 2A. The detailed information of distillation columns and the stream specifications of scenario 1 is demonstrated in Tables 1 and 2, respectively. In scenario 1, 60% of the energy consumption is reduced compared with the base case by applying the column heat integration system on the three-column Othmer distillation system.

More specifically in scenario 1, the first column of the three-column Othmer distillation system, named beer column (T-1), produces 57.2% (w/w) bioethanol as top-product under the vacuum pressure of 0.3 atm. Meanwhile, the bottom-product of the beer column is also sent to the stillage evaporation unit. In order to remove volatile components, the top-product of the beer column is conveyed to the second column (T-2) with an operating condenser pressure of 0.8 atm. The second column's top product, containing predominantly bioethanol (75% w/w), is sold as a by-product for the production of disinfectants (stream 3). The second column's bottom product, having almost 55% w/w bioethanol, is fed to the rectifier (T-3) under operating condenser pressure of 1.8 atm. The rectifier is the most significant column, as it has to prevent azeotropic mixture formation and purify bioethanol to 94%w/w, which is further purified through the dehydration unit. In the dehydration unit, pure bioethanol (stream 9) is produced using PSA with 4.8 molecular sieve adsorbent. A packed column, named scrubber, was used to recover the low amount of ethanol in the vent stream coming from the fermenters. Water is added on the top of this column to separate bioethanol from the vapor phase. After adding water to the scrubber, ethanol with a concentration of 2.2%(w/w) was conveyed to the beer column, T-1.

Except for scrubber, all columns were considered as trayed columns. Sieve-type trays were chosen due to their low price, high capacity, and low-pressure drop. Trayed-column diameter was

calculated with regard to flooding, foaming, and weeping factors. Furthermore, tray efficiency was assumed to be 60% based on data from Murphree, O'Connell's correlation, Van Winkle's correlation (for binary systems), and ALChE [29,42].

In the stillage evaporation unit of scenario 1, steam (stream 7) and vinasse (stream 8) were produced by a double-effect evaporator (Fig. 2A and Table 2). Evaporators were modeled through the combination of a shell and tube exchanger and flash vessel. In the tube side of the heat exchanger, the stillage was conveyed for ease of cleaning. Tube patterns in evaporators were set as triangular due to their higher heat-transfer rates. Evaporators were fabricated mainly of stainless steel 304. The considered allowable pressure drop is dependent on the fluid phase, viscosity (for liquids), and pressure (for gases and vapors). An allowable pressure drop of 0.5 atm was used for the cold side of two evaporators. Although the prediction of fouling factors is hard and they are usually extracted from former experiences, fouling factors of 0.00025 and 0.0002 were assumed for the steam and organic liquid, respectively [43,44].

3.2. Scenario 2

As shown in Fig. 2B, the second column (T-2) and rectifier (T-3) were coupled and integrated thermally to reduce the number of reboilers and energy consumption in scenario 2. More specifically, the heat duty of the second column reboiler should be provided by the latent heat of the rectifier side stream. To this intent, a vapor side stream, 41 kg/h, was taken from the rectifier to supply the second column reboiler's heat duty, 22.9 kW. Moreover, columns were integrated thermally for further energy efficiency. Regarding column pressures, reboilers and condensers were matched through cold and hot streams [45]. In scenario 2, the columns' energy demand with heat integration was 54% less than the columns without heat integration (Table 3). As compared with scenario 1, the total duty of distillation columns is higher in scenario 2, which may be the result of using a side stream instead of a reboiler. However, the use of thermally-coupled columns significantly reduces the capital cost. Accordingly, the production cost of ethanol in scenario 2 is 15% less costly than scenario 1.

3.3. Scenario 3

As illustrated in Fig. 2C, the dividing-wall column for the second column and rectifier was used to decrease the energy consumption and expenses in scenario 3. In this design, the second and third columns were converted to one column within a dividing-wall column. Since various separations are carried out on each side of the column, the column's dividing-wall was insulated to prevent heat transfer across the column. The pressure at both sides of the dividing column should be the same to operate in the industry. To this purpose, the second column's pressure was increased to 1.8 atm, which was the condenser operating pressure of the rectifier column (Table 3). Due to the existence of the rectifier side-stream, ethanol recovery in this column, 77% w/w, was not as large as other columns. Approximately 23%w/w of bioethanol was sent to the side stream, which was sold out as a by-product.

Furthermore, heat integration was applied to the columns for the reduction of energy requirement by 43%. As presented in Table 1, energy consumption in scenario 3, dividing-wall column together with heat integration, was reduced by 17.9% and 67% compared with scenario 1, the base case with and without column heat integration, respectively. The results showed that using a dividing-wall column could play an essential role in reducing distillation columns' energy consumption. Furthermore, dividing-wall columns have been also highly recommended for the

purification of lignocellulosic bioethanol production [46].

3.4. Scenario 4

In scenario 4, the pervaporation module was implemented as an alternative method to pressure swing adsorption in the dehydration unit. This module is a high potential process for separating azeotropic mixtures. During the pervaporation process, the process stream temperature decreases to provide the latent heat required for evaporation of the compounds. This reduced temperature results in a decrease in the transmembrane flux and increased membrane surface area. Therefore, a multi-stage pervaporation process was assumed due to significant cost reduction in the total required membrane areas. To improve the performance of multi-stage pervaporation, a heat exchanger was designed between every stage to compensate for the temperature drop of liquid feed during each step. A sensitivity analysis was performed using different membrane areas and energy requirements of the single-stage and multi-stage pervaporation process. The most efficient state of the pervaporation process was identified by implementing various operating conditions, namely, temperature, liquid feed composition, and the number of stages. Considering safe margin temperatures of PERVAP2210 and fixing the feed concentration at 5% (w/w) water, 85 and 90 were selected as the module operating temperatures.

According to the results (Table 4), the pervaporation process with an inlet concentration of 5% (w/w) water requires almost half of the area at 90 °C compared with 85 °C. This difference is due to the reduction in permeation flux as the temperature decreases. Moreover, two inlet water concentrations of 5% (w/w) and 10% (w/w) were fed to the module. The new feed concentration, 10% (w/w) water, results in a more significant energy-saving system. Considering the process's economy, four-stage pervaporation with 10% (w/w) water inlet at 90 showed the best outcomes.

In scenario 4, the four-stage pervaporation, having 10% (w/w) water inlet at 90 °C, and distillation system was integrated thermally for better efficiency (Fig. 3). In this scenario, the rectifier purifies bioethanol to 90% (w/w). While in the base case and scenario 1; bioethanol should be concentrated to 94% (w/w) to be conveyed to pressure swing adsorption. Consequently, scenario 4 contributes to 66% and 16.1% energy-saving compared with the base case and scenario 1, respectively. The detailed information on pervaporation modules in scenario 4 is presented in Table 5.

3.5. Scenario 5-9

In scenarios 5, 6, 7, 8, and 9, application of a single-effect, double-effect, and triple-effect evaporators without and with TVR system were included in the stillage evaporation unit. Natural gas and diesel are the most common fuels for steam production in small-scale plants in Iran [32]. Since steam costs are lower than

Table 5

Detailed results corresponding to the design of the pervaporation module in scenario 4 (cf. Fig. 3).

Pervaporation stages	1	2	3	4
Area (m ²)	9.0	9.0	9.0	9.0
Retentate (kg/hr.)	246.2	236.8	230.9	227.0
Water % (w/w)	5.9	3.0	1.3	0.5
Ethanol % (w/w)	94.1	97.0	98.7	99.5
Temperature (K)	346.8	352.4	357.5	361.0
Permeate (kg/hr.)	13.8	9.4	6.0	3.8
Water wt %	83.1	79.0	68.0	48.5
Ethanol wt %	16.9	21.0	32.0	51.5
Heat exchanger duty ^a (kW)	−0.9	3.7	2.4	1.2

^a Duty of the cooler or heater, situated before each stage.

electricity in Iran, which is due to the low price of energy carriers, the TVR system is proved to be a reasonable choice to be implemented for the improvement of thermal efficiency of the evaporators.

In the TVR system, 50% of the steam produced by the evaporator was recompressed by a steam jet venturi (nozzle-diffuser) and returned to the evaporator (Fig. 4). More specifically, motive steam (stream 2) at high pressure (10 atm) expands as it passes through the diffuser. In the diffuser, recycled vapor was entrained and discharged at an intermediate pressure to the evaporator. The detailed specifications of the streams in scenario 6 are presented in Table 6.

Among different scenarios in the stillage evaporation unit, scenario 6 demonstrated the best economic performance. In scenario 6, the investment costs cut by 30% compared with the double-effect evaporators without a TVR system in the base case. Even though the fresh steam requirement was increased in this scenario, the considerable reduction in capital costs provides the lowest bioethanol production price.

3.6. Summary of scenarios 1-9

Based on the results presented in Table 7, the proposed scenarios in separation and dehydration units, scenarios 1, 2, 3, and 4 reduced energy consumption and total capital costs, and thus enhancing the process's economic profitability. The internal rate of return was increased from 22.0% in the base case to 22.1, 31.0, 27.2, and 26.0% in scenarios 1, 2, 3, and 4, respectively. Because of lower operating and

Table 6

Stream specifications of stillage evaporation unit for scenario 6 (cf. Fig. 4.).

Streams	1	2	3	4
Flow rate (kg/h)	2491.6	1548.0	2745.0	2393.9
Water (%w/w)	97.0	100.0	100.0	99.0
Temperature (K)	349.5	453.1	451.3	336.9
Pressure (atm)	0.4	9.9	9.9	0.2

Table 4

The effect of temperature, feed composition, and stage number in pervaporation module on the energy demands and required surface area of dehydration unit for scenario 4.

Temperature	Water at inlet stage (% w/w)	Stage number	Area (m)	Rectifier duty (kW)	Cooler duty (kW)	Heat exchanger duties (kW)	Total duty ^a (kW)
85	5.0	1	394.5	828.8	−2.1	—	830.9
85	5.0	3	51.0	828.8	−2.1	3.1	834.0
85	5.0	4	44.0	828.8	−2.1	3.6	834.5
85	5.0	5	45.0	828.8	−2.1	4.1	835.0
90	5.0	1	142.0	828.8	−0.9	—	830.9
90	5.0	3	27.0	828.8	−0.9	3.3	833.0
90	10.0	1	371.5	313.8	−0.9	—	314.7
90	10.0	3	45.0	313.8	−0.9	6.8	321.5
90	10.0	4	36.0	313.8	−0.9	7.3	322.0

^a Total duty includes the rectifier's duty, cooler situated before the pervaporation module, and heater duties located between each stage.

Table 7

A summary of base case and nine scenarios along with the results of profitability analysis.

Parameters	Base case	Scenario								
		1	2 ^g	3 ^g	4 ^g	5	6	7	8	9
Internal rate of return (%)	22.0	22.1	31.0	27.2	26.0	29.3	39.9	35.1	25.5	31.5
MIRR ^a (%)	20.0	20.1	21.8	21.1	20.9	21.5	23.1	22.5	20.8	21.9
Payout period (year)	9.0	9.0	6.4	7.4	7.8	6.8	4.9	5.6	8.1	6.3
Profitability index	1.00	1.1	1.16	1.10	1.08	1.14	1.3	1.23	1.07	1.18
Capital costs (million US\$) ^b	6.4	6.4	5.3	5.2	5.6	5.1	4.5	4.8	5.8	5.1
Total production cost of ethanol (US\$/L)	0.53	0.52	0.44	0.50	0.50	0.54	0.35	0.43	0.52	0.44
CHI ^c	No	Yes	Yes	Yes	Yes	No	No	No	No	No
TVR ^d	No	No	No	No	No	No	Yes	Yes	No	Yes
Total duty ^e (kW)	1200.5	482.5	608.9	396.2	404.7	1200.5	1200.5	1200.5	1200.5	1200.5
Fresh steam ^f (kg/hr.)	1335.0	1335.0	1335.0	1335.0	1335.0	2745.0	1548.0	1009.0	888.0	762.0

^a MIRR: Modified internal rate of return.^b The presented capital cost corresponds to the total capital costs including feed handling, fermentation, separation, dehydration, and evaporation.^c CHI: Column heat integration, applied to the separation and dehydration unit.^d TVR: Thermal vapor recompression is used in the stillage evaporation unit.^e Total duty of the separation unit.^f Required fresh steam for the stillage evaporation unit. In scenarios 1–4, a falling film evaporator followed by a forced circulation evaporator was used in the stillage evaporation unit.^g For scenarios 2, 3, and 4, the total energy for the process without heat integration is presented in Table 3.

capital costs, the payout period was decreased from 9.0 in the base case to 6.4, 7.4, and 7.8 years for scenarios 2 to 4, respectively. Notwithstanding the reduction in ethanol's total production cost due to the decrease of energy demands, scenario 1 failed to appreciably change the payout period compared with that of the base case. Among scenarios 1 to 4, scenario 2 showed the best financial results in the separation and dehydration unit. Although in terms of capital costs, scenario 2 was more expensive than scenario 3, better economic features were attained in scenario 2. Ethanol could be at a reasonably same price in scenarios 3 and 4; however, scenario 3 had a better payout period and profitability index due to the lower capital costs.

Besides the separation and dehydration unit, the stillage evaporation unit was considered in scenarios 5 to 9. In scenarios 6, 7, and 9, the superior profitability index could be obtained through the TVR system (Table 7). In the TVR scenario, the temperature driving forces were increased, which resulted in the reduction of required heat exchanger surface areas. Consequently, lower investment costs were needed in the TVR scenario. Using the TVR system, double- and triple-effect evaporators were compared with single-effect evaporator, which reduced capital costs from 5.1 to 4.8 in scenarios 7 and 9 to 4.5 million US\$ in scenario 6. Therefore, in scenario 6 the bioethanol production cost was decreased from 0.53 in the base case to 0.35 US\$/L. Scenario 6 showed the best economic features concerning capital and operating costs.

4. Sensitivity analysis

Sensitivity analysis was conveyed to investigate the influence of the most affecting parameters and nine suggested scenarios on bioethanol total production cost. Total production cost was divided into four groups: direct production costs, fixed charges, plant-overhead costs, and general expenses [47]. For instance, direct production costs contained several items, namely, raw material, operating labor, direct supervisory and clerical labor, utilities, maintenance, and repairs as well as operating supplies.

In Fig. 5, the raw material sector, consisting of water, molasses, and yeast, which accounted for the highest total production cost, is presented. Since yeast was consumed far less than water and molasses in the bioethanol production process, water and molasses showed the highest impact on ethanol production cost (Fig. 6). In other words, molasses and water price fluctuations had an immense bearing on the bioethanol price.

The bioethanol total production costs for scenarios 1 to 9 is presented in Fig. 7. The highest ethanol production cost belonged to the base case, with 0.533 US\$/L, while implementing different scenarios reduced the production cost to 0.35–0.52 US\$/L. The lowest ethanol prices, 0.44 and 0.35 US\$/L were achievable by applying thermally-coupled column in the separation and dehydration unit (scenario 2) and a single-effect evaporator with TVR system (scenario 6) in the stillage evaporation unit. Concerning the lower heating values of ethanol (21.2 MJ/L) and gasoline (32.0 MJ/L) as well as the density of ethanol, the gasoline-equivalent price was calculated as follows:

Gasoline equivalent price (US\$/L) = Bioethanol production price (US\$/kg) * ethanol density (kg/L) * gasoline LHV/ethanol LHV.

From Fig. 7, bioethanol, with minimum fossil fuel burning and GHG emission, had a lower minimum selling price than gasoline. Thus, a higher FER with lower production cost could be obtained through the bioethanol production process in all scenarios, 1 to 9.

5. Comparison with other processes

The production process of bioethanol was optimized for energy-consuming and costly units, namely, separation, dehydration, and stillage evaporation units. Coincidence analysis of the mentioned units distinguishes this research from others. Although several studies had been conducted to lower the energy demand and costs, the research areas were predominantly limited to the dehydration unit, using extractive or azeotropic distillation. Therefore, only the dehydration unit should be considered in order to compare the results of this study with others. The comparison of the results is presented in Table S3.

In this research, the bioethanol dewatering unit profits from a four-stage pervaporation module. The four-stage pervaporation module reduced over 60% energy consumption and 70% capital costs, while the best results among former investigations using extractive distillation, presented by Figueirêdo et al. [48], lowered energy requirement and expenses by 17.9% and 21.9%, respectively.

6. Conclusions

This study presents economically viable strategies for further energy reductions in a commercial case-study for ethanol production. The most energy demanding units with high operating costs of bioethanol production plant are separation, dehydration,

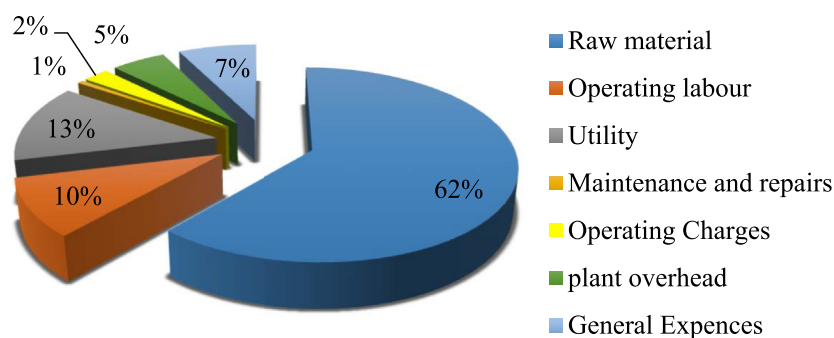


Fig. 5. Contribution of different parts in total production cost of bioethanol for the base case.

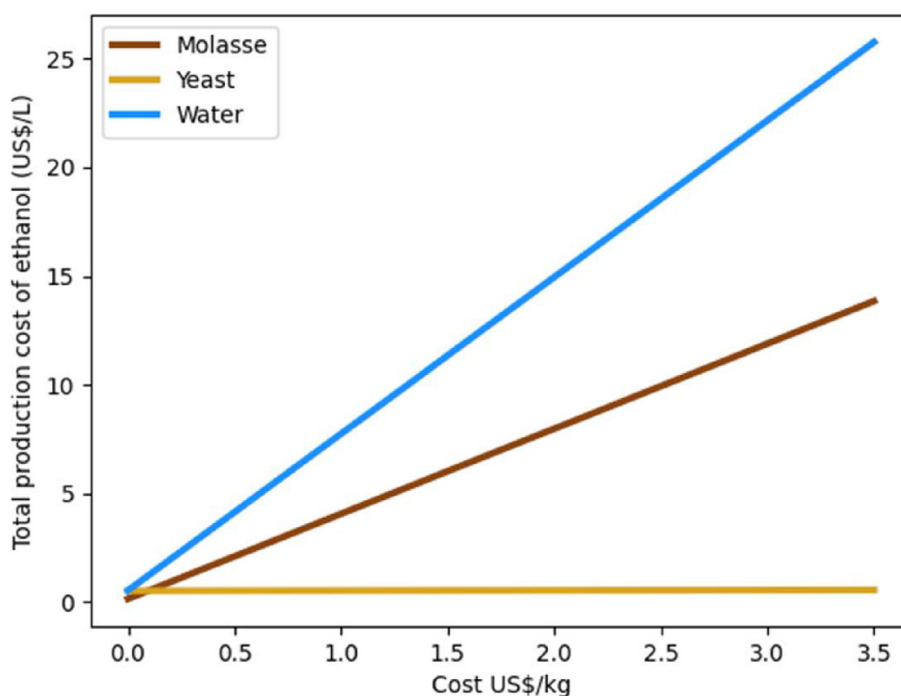


Fig. 6. The effect of raw material price on the total production cost of bioethanol.

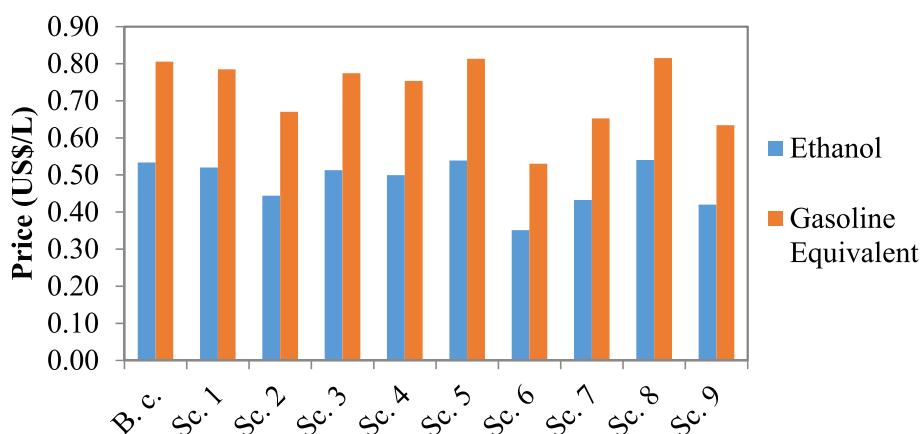


Fig. 7. Gasoline equivalent price of biofuels as well as the minimum ethanol selling price for all scenarios.

and stillage evaporation units. In this study, nine scenarios in the mentioned units were investigated for enhanced thermal and

economic efficiency.

Based on the results, the bioethanol separation system benefited

from employing thermally-coupled columns and dividing-wall column and heat integration. Thermal coupling of columns reduced the process energy requirement and capital costs by 49% and 17%, respectively. In contrast, the application of the dividing-wall column saved 67% of energy demand, and 19% of expenses compared with the base case. The base case consists of a conventional three-column separation system without column heat integration. The results indicated that applying the mentioned methods and column heat integration plays an essential role in optimizing the separation system.

The multi-stage pervaporation module handled the azeotropic mixture among different bioethanol dehydration methods and resulted in the highest FER and higher profitability index. In contrast to the base case, the four-stage pervaporation module reduced energy consumption by 60% and capital costs by 70% decrease. The optimized four-stage pervaporation module seems to be an efficient alternative method to pressure swing adsorption for bioethanol dewatering.

Regarding the stillage evaporation unit results, the thermal vapor recompression (TVR) system lowered the investment costs by 30%, while no external energy source was required in its mechanism. Considering fresh steam demand and expenses, a single-effect evaporator with TVR system presented more reasonable results than the double-effect evaporator without TVR in the base case.

Sensitivity analysis revealed that bioethanol's total production costs decreased from 0.53 US\$/L to 0.35–0.50 US\$/L for scenarios 1–9, while the average gasoline (95 unleaded) price in the Middle East was 0.39 US\$/L in late 2016 [49]. The gasoline price was increased to 0.51 US\$/L in 2018. Thus, the ethanol production process's energy consumption and ethanol production costs improved through new practical methods.

Credit author statement

Maryam Botshekan: Software, Formal analysis, Investigation, Data Curation, Visualization Writing - Original Draft. **Ahmad Moheb:** Supervision, Project administration, Writing - Review & Editing. **Fatemeh Vatankhah:** Methodology, Software, Investigation, Data Curation, Formal analysis. **Keikhosro Karimi:** Conceptualization, Project administration, Supervision, Project administration, Funding acquisition, Writing - Review & Editing. **Marzieh Shafiei:** Supervision, Visualization, Software, Methodology, Project administration, Funding acquisition, Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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