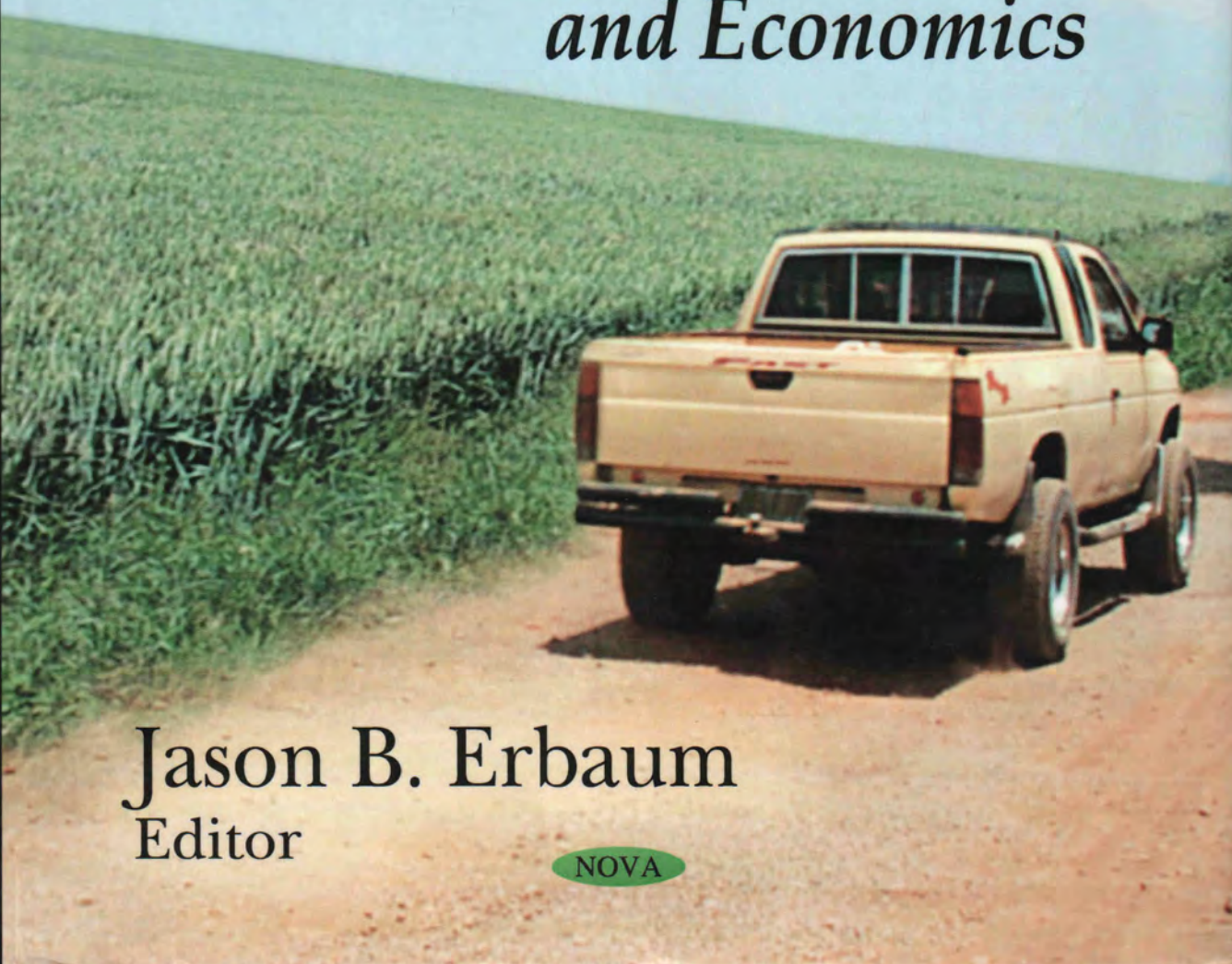




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*Production, Benefits
and Economics*



Jason B. Erbaum
Editor

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**BIOETHANOL: PRODUCTION, BENEFITS
AND ECONOMICS**

JASON B. ERBAUM
EDITOR

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Chapter 6

ANALYSIS OF ENERGY CONSUMPTION OF DISTILLATION OPTIONS TO OBTAIN HIGH-PURITY BIOETHANOL

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ABSTRACT

Among biofuels, bioethanol has increased in importance in many countries because it can be used directly or mixed with gasoline in combustion engines. The production of bioethanol in a fermentative process usually gives a dilute solution from which the bioethanol must be obtained in a high concentration in order to be used as biofuel. The use of bioethanol mixed with gasoline in combustion engines is associated with fewer emissions of both hydrocarbons and carbon monoxide. The production of high purity bioethanol using extractive distillation sequences with ethylene glycol or a dilute solution of NaCl as entrainers are studied in detail in terms of energy consumption and total

annual costs. Conventional and complex distillation sequences are designed optimally in a computational framework implemented in Aspen PlusTM and MatlabTM. The results indicate that complex distillation sequences involving thermal linking can reduce energy consumption over conventional distillation sequences using either ethylene glycol or a dilute solution of NaCl as entrainers. As a result, significant reductions in total annual cost can be obtained in the production of high purity bioethanol. This can position ethanol as a competitive biofuel when compared to gasoline.

INTRODUCTION

A large portion of the world economy has been based on processes highly dependent on petroleum; as a consequence, two significant problems have been created: the reduction of oil reserves, and the increase in gas emissions that has been associated with global warming. Researchers in many areas are working on solutions to mitigate these problems. One option that contributes a partial solution is intensification in the production and use of biofuels, including biodiesel, bioethanol, biomass, etc. Of these options, bioethanol is being currently used in combustion engines without modifications in a mixture of up to 20 %. The use of this mixture is important because it allows improved oxidation of hydrocarbons and, as a result, reduction in both hydrocarbons and carbon monoxide emissions (Quintero et al., [1]).

Currently, most bioethanol production is obtained from sugar cane, and, secondly, from corn, although research efforts are focused on industrial production from lignocellulosic material such as agricultural and forest residues. In the production process, four main steps can be identified: Treatment of the raw material to obtain cellulosic mass, saccharification to obtain sugars from the cellulosic mass, fermentation of the sugars, and recovery of ethanol. Independently from the raw material and/or the process used, the product obtained from the fermentation step is a dilute solution of ethanol in water, from which ethanol is separated and purified to the desired concentration. In addition to the research effort in the saccharification and fermentation process, the separation step must also be viewed as a challenge because of the energy that it requires. Assuming that the fermentation process produces a dilute solution of ethanol in water (10 % in moles of ethanol) that requires treatment in order to obtain high purity ethanol that then can be mixed with gasoline [2,3], the production of high purity ethanol using distillation requires significant quantities of energy and mass separation agents such as ethylene glycol, NaCl, KI, or CaCl₂.

Distillation is widely used for the separation of many fluid mixtures, but this separation option presents two disadvantages: its high energy consumption and low thermodynamic efficiency (Hernández et al., [4]). Taking into account that distillation is the most important separation option in many industries, researchers and process engineers have been working in order to improve distillation sequences. Two approaches have been used: heat integration in distillation sequences and thermal linking of distillation columns of the sequence. By using these two techniques, important energy savings can be obtained. For instance, in the case of thermally coupled distillation options, the energy savings obtained are in the range of 30 to 50 % over distillation schemes based on conventional distillation columns for the separation of multicomponent mixtures [5-8]. In the case of ternary mixtures (A,B,C), the two conventional distillation sequences are depicted in Figure 1. Figure 1a is the so-called direct distillation sequence in which the components are removed as overhead products, and the distillation

sequence shown in Figure 1b is the indirect distillation sequence that recovers the components as bottoms products. It has been reported by Tedder and Rudd [9] that these conventional distillation options are the most appropriate for the separation of mixtures with high contents of either the lightest or heaviest component respectively.

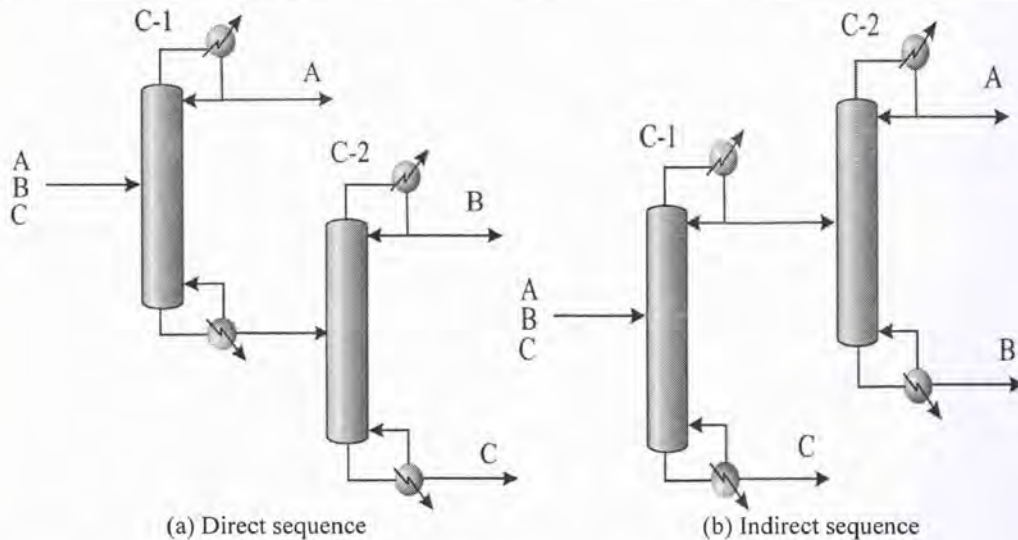


Figure 1. Conventional distillation sequences for ternary separations.

Alternatively, the sequences including thermal links (Figure 2) can be good options for the separation of mixtures with low or high content of the intermediate component. There are many thermally coupled distillation sequences, but those indicated in Figure 2 have been studied in greater detail and some practical implementations have been reported. The thermally coupled distillation sequence indicated in Figure 2a (TCDS-SR) requires a main distillation column coupled to a side rectifier by liquid and vapor streams. This complex distillation option offers energy savings of up to 30% over the conventional direct distillation sequence for the separation of mixtures with less than 15% of the intermediate component in the feed and if the split A/B is easier than B/C. For the same content of the intermediate component, and when the split B/C is easier than A/B, the TCDS-SS (Figure 2b) is the best option. Moreover, the complex distillation option shown in Figure 2c can lower energy consumption up to 50% for the separation of mixtures with high content of the intermediate component in the feed. The last thermally coupled distillation option is called the Petlyuk distillation column; this complex distillation sequence has been implemented in industrial practice by using a dividing wall distillation column. This option uses a single shell divided by a wall (Figure 3), and savings have been reported in both energy and capital costs.

It is important to highlight why the thermally coupled distillation sequences can be better options than conventional distillation sequences for some separations. The explanation can be formulated in terms of the profiles of the intermediate component in the first distillation column of the conventional direct distillation sequence. The composition profile of the intermediate component reaches a maximum below the feed stage and then diminishes as the bottoms part is reached (Figure 4). This effect is known as remixing and is associated with higher energy consumption, because additional energy is required in the second distillation

sequence to re-purify the mixture (Triantafyllou and Smith, [10]). A similar effect is observed in the composition profile of the intermediate component in the indirect distillation sequence, but this maximum is reached above the feed stage. This important aspect is taken into account in the design of the dividing wall distillation column, because the side stream is taken from the stage where the composition of the intermediate component presents the maximum.

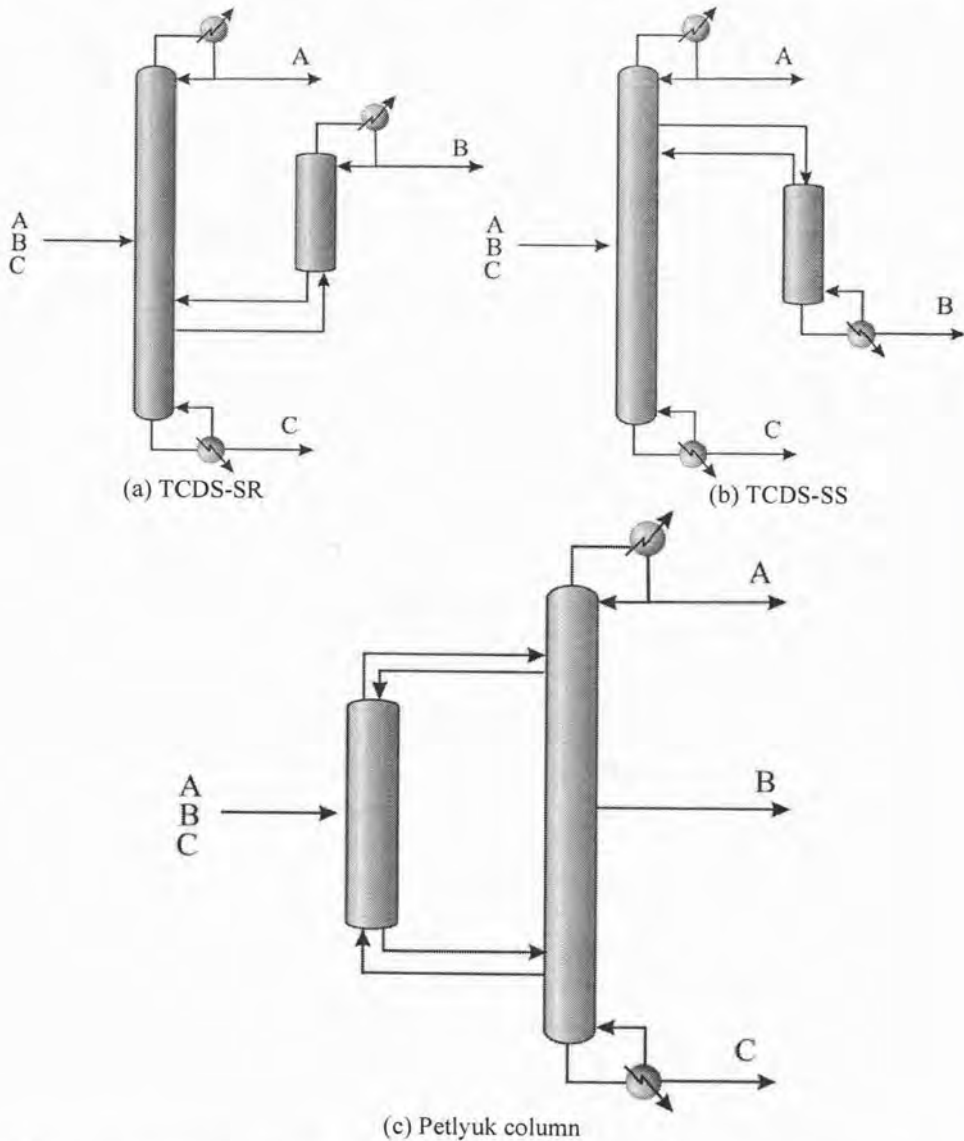


Figure 2. Thermally coupled distillation options for ternary separations: (a) Direct thermally coupled distillation sequence (TCDS-SR), (b) Indirect thermally coupled distillation sequence (TCDS-SS), (c) Petlyuk distillation column.

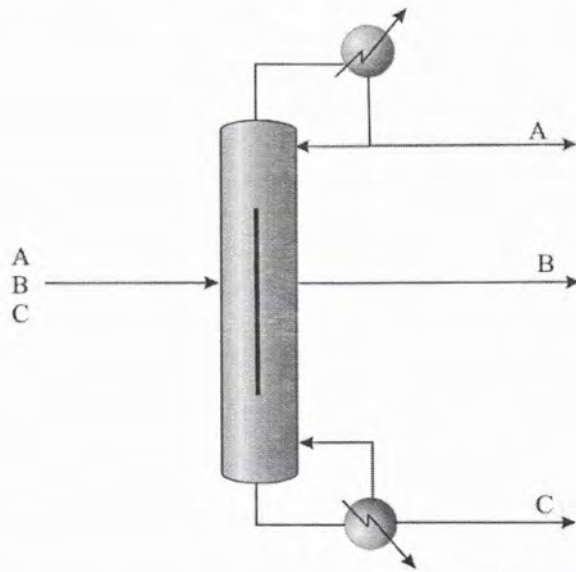


Figure 3. Dividing wall distillation column for ternary separations.

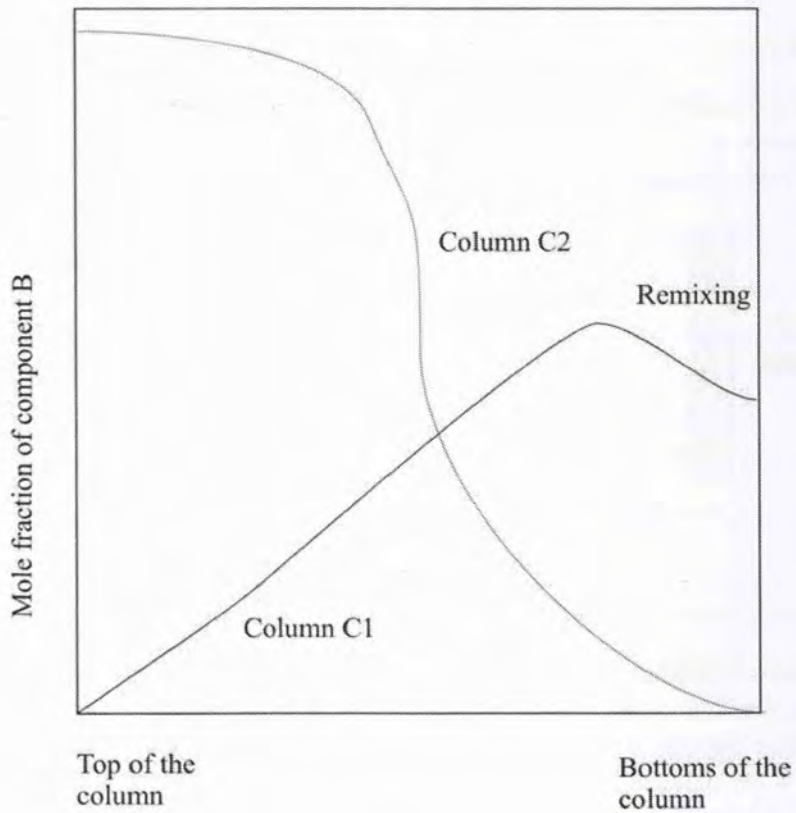


Figure 4. Composition profiles of the intermediate component for the conventional direct distillation sequence.

PROBLEM STATEMENT

As indicated in the preceding section, we are interested in comparing the optimal energy consumption and total annual cost of a several schemes for obtaining high purity ethanol from a dilute solution (10% in moles of ethanol in water). The dilute solution is introduced into a distillation column in order to obtain a distillate product with composition nearly at azeotropic condition (96% in mass of ethanol in water), and almost pure water as bottoms product. The distillate product is introduced into an extractive distillation column using either ethylene glycol or a solution of NaCl as mass separation agents (entrainers). This second stage produces high purity ethanol with a mass fraction of 0.995 and the mass separation agent is recovered. The following distillation options are designed and compared in terms of energy consumption.

The first option is the conventional distillation sequence requiring an additional distillation column using ethylene glycol as entrainer to obtain ethanol as distillate product (mass fraction of 0.995) and a binary mixture of entrainer and water as bottoms product. The bottoms product is introduced into a distillation column to recover the entrainer as shown in Figure 5.

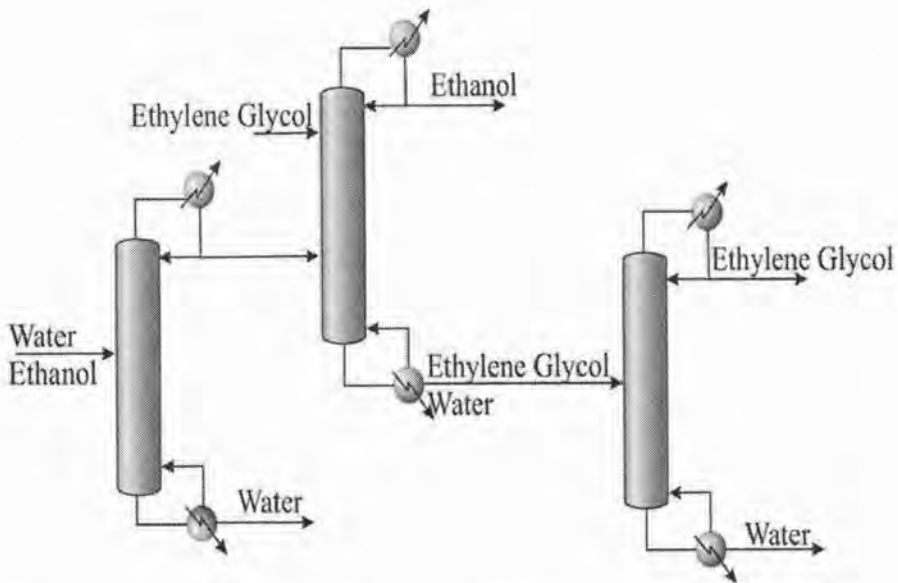


Figure 5. Extractive conventional distillation sequence (ECDS).

The second distillation sequence is the thermally coupled distillation sequence with a side rectifier. The distillate of the main column is ethanol with the required purity and the bottoms product of this column is the entrainer. The side rectifier column removes a mixture of ethanol and water that can be recycled to the first distillation column. The distillation option is described in Figure 6; as can be noted, this complex distillation option includes a recycle stream from the side rectifier to the main distillation column. This recycle stream plays an important role in the energy efficiency of the distillation scheme, and it is necessary to detect the value that corresponds to the minimum energy consumption.

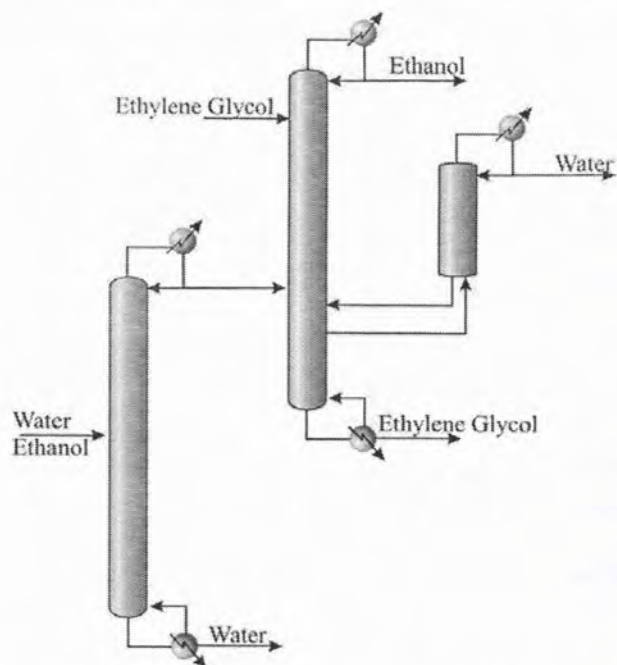


Figure 6. Extractive thermally coupled distillation sequence with a side rectifier (ETCDS-SR).

The third scheme analyzed involves the use of a distillation column with a side stream. In this case, the distillate product is high purity ethanol, the side stream is a dilute mixture of ethanol and water that can be recycled to the azeotropic distillation column and the bottoms product is ethylene glycol (Figure 7).

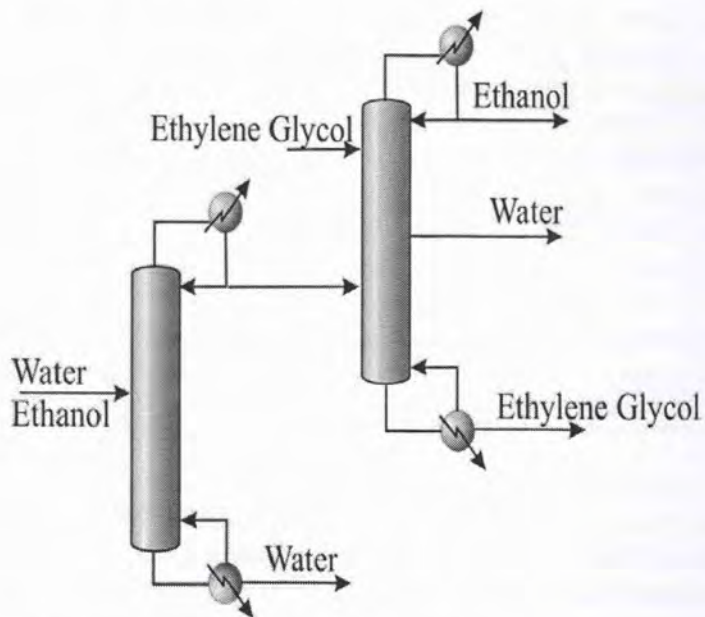


Figure 7. Extractive distillation sequence with a sidestream (EDSS).

The fourth option includes extractive dividing wall distillation sequences (one or two walls). These options recover the ethanol in the distillate and the bottoms product is the entrainer. The sidestream removes a mixture of ethanol and water that can be returned to the first conventional distillation column. These two options are schematically shown in Figures 8 and 9, respectively. These options include two or four recycle streams that must be varied in order to obtain the minimum energy demand in the reboiler. It is important to highlight that these fully thermally coupled distillation sequences can be implemented in industrial practice [11] by using one or two dividing walls as depicted in Figures 8 and 9 respectively.

DESIGN AND OPTIMIZATION PROCEDURES

In order to guarantee minimum energy consumption in the distillation options, an optimization procedure based on genetic algorithms was implemented in Aspen PlusTM and MatlabTM. This computational framework is required because many search variables are involved in the optimization problem. These variables can be continuous or discrete.

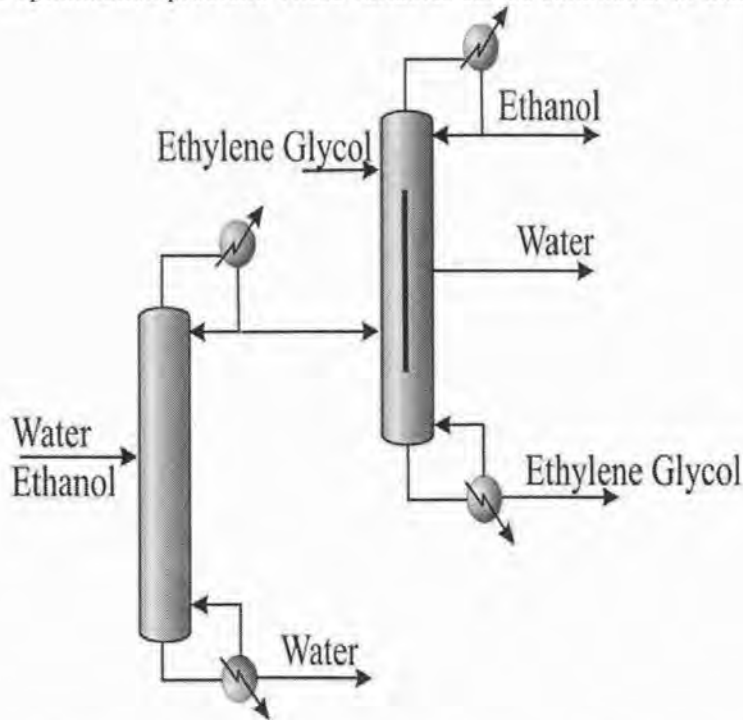


Figure 8. Extractive distillation sequence with one dividing wall (EODWDC).

The operational pressure was not considered a search variable, since it was set in order to guarantee the use of cooling water in all condensers. All condensers are operated at 14.7 psia using cooling water. Furthermore, it is important to mention that in all cases, the constraint for the composition of ethanol was a mass fraction of 0.995.

The first distillation column used in all the distillation schemes has the minimum number of search variables, but we have continuous and discrete variables in the optimization procedure; for instance, the number of actual stages, feed stage, reflux ratio and distillate rate.

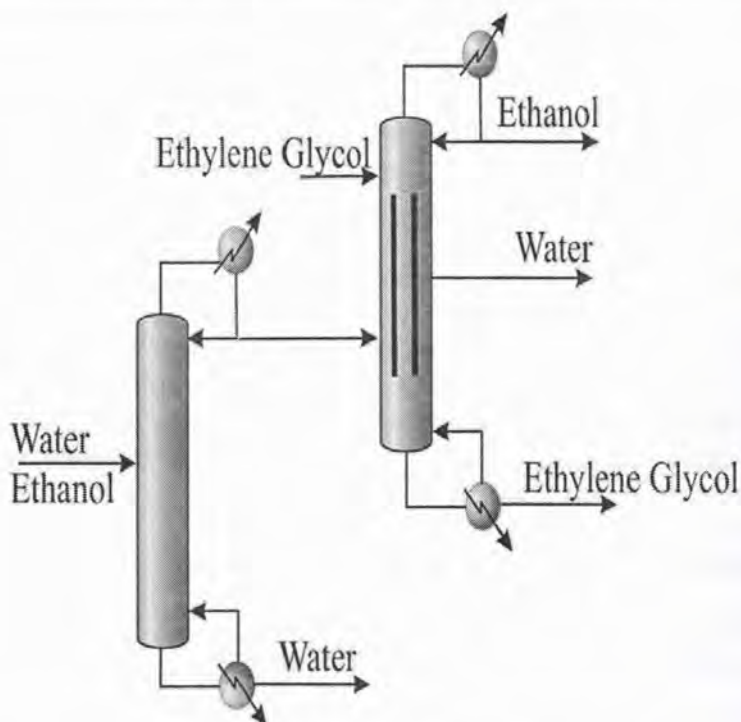


Figure 9. Extractive distillation sequence with two dividing walls (ETDWDC).

The second distillate column indicated in Figure 5 includes two additional search variables, the feed stage of the entrainer, and the ratio of flows of the entrainer to the feed stream. The procedure for optimal design of the thermally coupled distillation column included in Figure 6 is truly a difficult task, because the recycle stream couples the main distillation column and the side rectifier. This implies that the designs of the columns must be obtained simultaneously. The additional variables in this case are the positions of the liquid and vapor streams between the two columns and the flow of the interconnecting vapor stream.

When Figure 7 is analyzed, we detect important design variables for the second distillation option, for instance, the feed stages, the position and flow of the sidestream.

For the design of the fully coupled distillation options indicated in Figures 8 and 9, the procedure increases the complexity as the number of recycle streams increases; for instance, for one dividing wall there are two recycle streams, meanwhile for two dividing walls, the number of recycle streams increases to four. As we can see, obtaining the optimal designs of the distillation options is a difficult task to complete without the use of a computational framework.

COMPUTATIONAL FRAMEWORK

Obtaining the optimal designs of the extractive distillation sequences is a complex task, since it implies determination of integer and continuous variables. These variables combination must provide the minimal number of stages and heat duty in each column of the sequence. In addition, the purities required in each stream product must be satisfied.

In other words, finding an optimal design means solving a multiobjective, highly nonlinear problem with constraints. The problem is multiobjective because we know beforehand that the number of stages and the heat duty are variables in competition, since we cannot indefinitely decrease one without increasing the other. The problem is highly nonlinear because we are considering the complete set of MESH (component mass balances, equilibrium relationship, summation constraints, energy balance) equations along with the phase equilibrium calculations. Finally, purities and/or recoveries that have to be reached represent the constraints of the problem.

Table 1 presents the number of manipulated variables, along with the number of objectives to be minimized and the constraints considered for each distillation option.

The number of manipulated variables changes because it depends on the complexity of the structure considered; also, the number of reboilers and shells determines the number of objectives that have to be minimized simultaneously. Finally, the purities required are four since we have four product streams; in addition, three recoveries are considered as constraints.

Table 1. Distillation options and their manipulated variables, objectives and constraints

Distillation option	Manipulated variables	Number of objectives		Constraints considered	
		Integer and continuous	Number of stages	Heat duty	Purities
ECDS	14	3	3	4	3
ETCDS-SR	14	3	2	4	3
EDSS	12	2	2	4	3
EODWDC	15	3	2	4	3
ETDWDC	17	4	2	4	3

In order to optimize the conventional and thermal coupled extractive sequences we used the multiobjective genetic algorithm with constraints coupled to Aspen PlusTM developed by Gutiérrez-Antonio et al. [12]. Their algorithm manages the constraints using a multiobjective technique based on the concept of non dominance for constraints-handling proposed by Coello-Coello [13], which guides the NSGA-II search (Meyarivan et al, [14]). Since their code is coupled to commercial simulator Aspen Plus, all the results obtained here considered the rigorous energy and material balances, along with the equilibrium phase calculations.

For all sequences, we used 1600 individuals and 80 generations as parameters of the algorithm. These parameters were obtained through a tuning process, where we performed several runs of the algorithm with different number of individuals and generations.

RESULTS

As indicated above, high purity ethanol will be used pure or mixed with gasoline in combustion engines. For that reason, it is crucial that the composition reach the desired value (mass fraction of ethanol of 0.995) in the extractive distillation columns.

It is important to analyze the composition profiles of the optimal designs obtained in the computational framework. As depicted in Figures 5-9, the role of the first distillation column is the separation of most of the water in the bottoms product; according to Figure 10, the bottoms product is almost pure water obtained from stage number 30. Meanwhile, the top product is an enriched binary mixture of ethanol and water. This stream is sent to the second purification option consisting of extractive distillation schemes. First, the use of the ethylene glycol was analyzed, and the results are presented in Figures 11-13.

Figure 11 presents the composition profiles of the extractive distillation column of Figure 5, where it can be noted that the distillate product is ethanol with the required purity, but the bottoms product is a binary mixture of ethylene glycol and water. In this case it is important to highlight that an additional distillation column will be required to recover the ethylene glycol, and its energy consumption will be increased because the remixing effect is presented in the extractive distillation column. This inefficiency in the separation process is indicated in Figure 11; the composition of water increases until a maximum concentration at stage 18, but as the bottom of the distillation column is reached, the composition decreases drastically.

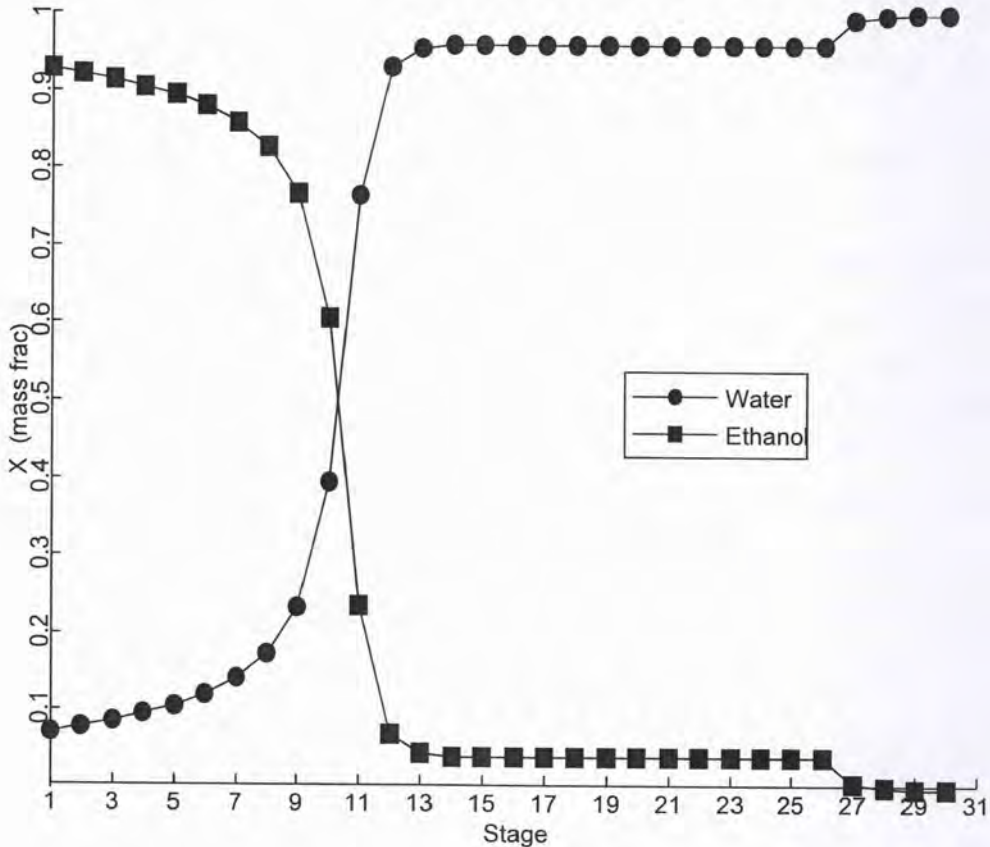


Figure 10. Composition profiles in the conventional distillation column.

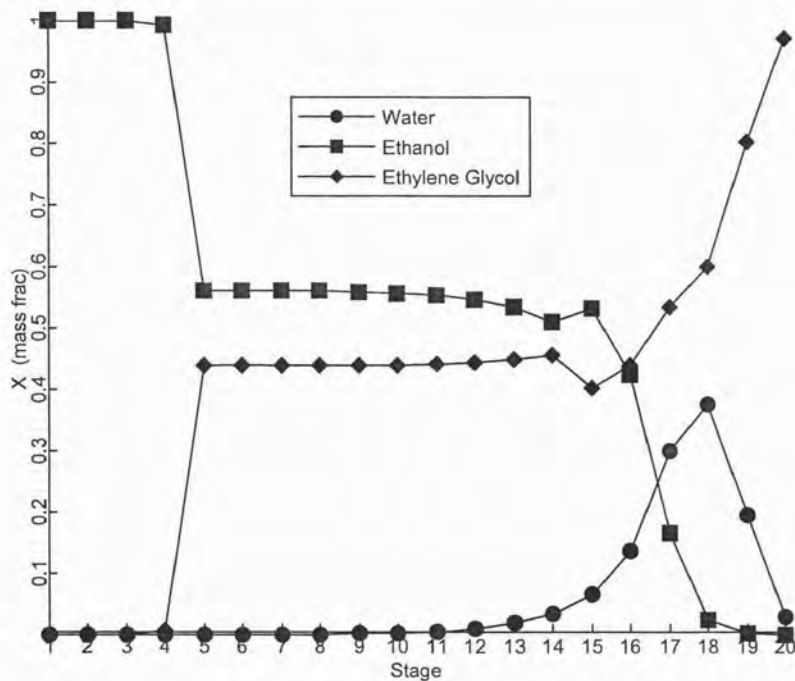


Figure 11. Composition profiles of the extractive distillation column.

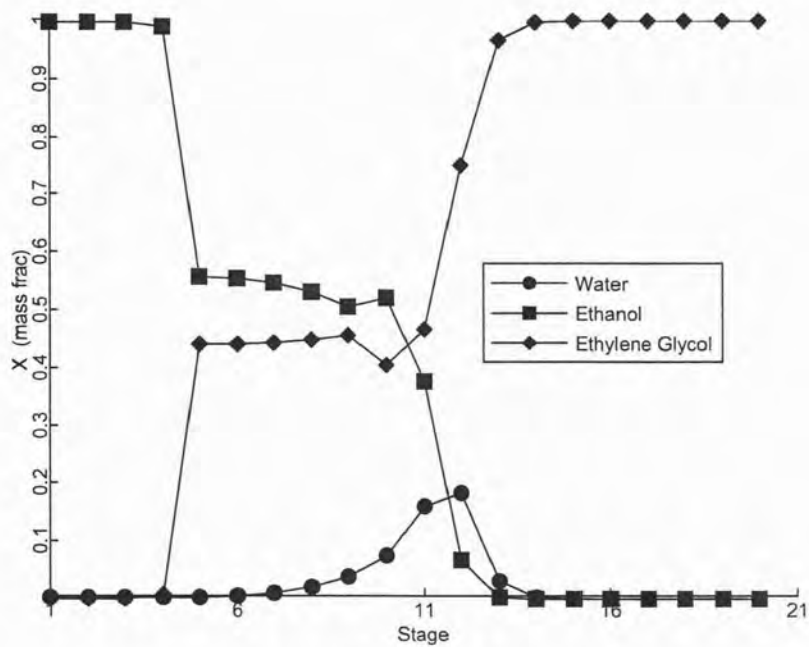


Figure 12. Composition profiles in the main column of the ETCDS-SR.

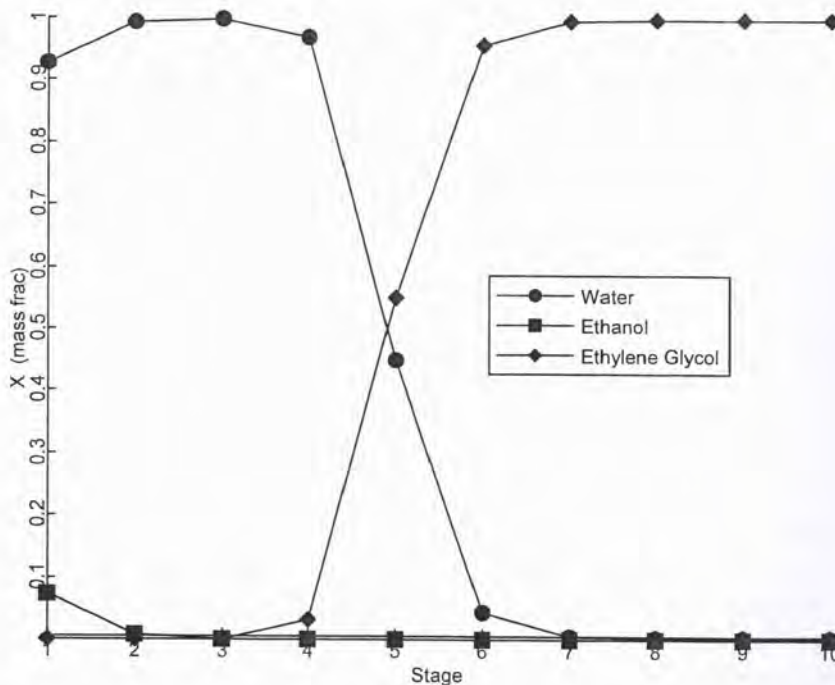


Figure 13. Composition profiles in the side rectifier of the ETCDS-SR.

In comparison with the separations achieved in the extractive distillation column of Figure 5, for the extractive complex distillation sequences of Figures 6-9, no additional distillation columns are required to recover ethylene glycol. To illustrate this, composition profiles of the extractive TCDS-SR are shown in figures 12 and 13 for the main and side rectifier columns, respectively. In the main distillation column (Figure 12), the top product is ethanol with the required purity, and the bottoms product is ethylene glycol. The side rectifier (Figure 13) has a dilute mixture of ethanol as top product that can be returned to the conventional distillation column (a distillation column that is common to the distillation sequences). The composition profiles for the distillation column with a sidestream and the distillation columns with one or two dividing walls are similar to those of the extractive ETCDS-SR, but the sidestream product is a binary mixture of ethanol and water that can be recycled to the first conventional distillation column. Again, the distillate is high purity ethanol and the bottoms product is the recovered ethylene glycol. According to the composition profiles of the distillation columns, where the ethylene glycol is used as entrainer, it can be concluded that this mass separation agent can be suitable for obtaining high purity ethanol since all the distillation sequences can adjust the composition to the desired value.

In order to test the other mass separation agents, the five distillation sequences were studied using a solution of NaCl with a mass concentration of 10%, but the composition of the ethanol in the product stream could not be achieved. For example, Figure 14 presents the composition profiles for the use of the solution of NaCl as entrainer; it can be noted that the composition of the product is around 0.98 mass fraction of ethanol. This value does not fit the

purity constraint. As a result, it can be concluded that saline extraction cannot be used to obtain high purity bioethanol. Considering these results, the energy consumption and total annual costs were calculated only for the use of ethylene glycol as entrainer.

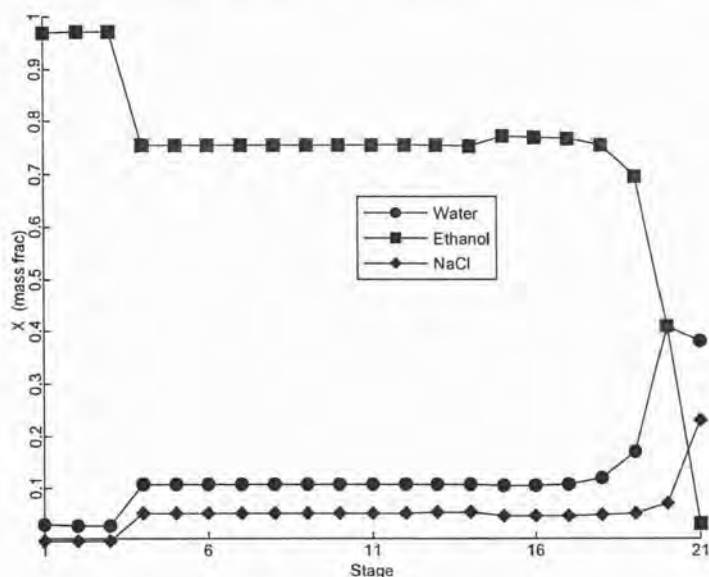


Figure 14. Composition profiles of the extractive distillation column.

The results regarding the energy consumption are shown in Table 2; it can be seen that the minimum energy consumption is presented by the conventional extractive distillation sequence. In addition, the extractive thermally coupled distillation sequence involving a side rectifier presents similar energy consumption. The other complex distillation sequences present energy consumptions that are 115% higher than those obtained in the best two options in terms of energy consumption (ECDS, ETCDS-SR). However, it is important to compare the extractive distillation sequences in terms of energy consumptions; this comparison does not take into account the temperatures of the utilities and the costs associated with the purchase, installation and operation of the extractive distillation sequences. In order to obtain a more realistic comparison, total annual costs (Equation 1) were obtained for the extractive distillation sequences (Table 2). According to the total annual costs, the best option is the extractive thermally coupled distillation sequence with a side rectifier, followed by the extractive distillation sequence with a sidestream.

$$\text{Total Annual Cost (TAC)} = \text{utility costs} + (\text{capital investment} / 5 \text{ years}) \quad (1)$$

Finally, in accordance with the total annual costs, the best option for purification of bioethanol is the extractive thermally coupled distillation sequence with a side rectifier. This result is important because the total annual cost can be reduced further using a dividing wall for an industrial implementation. Figure 15 presents the proposed extractive distillation sequence using a dividing wall distillation column.

Table 2. Total energy consumption (Q) and total annual cost (TAC) of the distillation sequences

Distillation sequence	Q (kW)	TAC (USD/year)
ECDS	314.8	291,553.41
ETCDS-SR	333.1	280,806.84
EDSS	524.2	387,228.90
EODWDC	607.9	390,373.64
ETDWDC	678.3	416,374.76

CONCLUSION

The process of obtaining high purity bioethanol from a dilute solution of a fermentation process was studied considering different extractive distillation sequences. The optimal designs of the extractive distillation sequences were obtained in a computational framework implemented in a commercial simulator and mathematical software using genetic algorithms. Ethylene glycol and a dilute solution of NaCl were considered to be mass separation agents in the extractive distillation sequences. Ethylene glycol was selected as a good option, since required purity of bioethanol could be achieved. Although NaCl is cheaper than ethylene glycol, this is not a suitable option because the composition of ethanol could not be adjusted to the desired value. The extractive distillation sequence with a side rectifier using ethylene glycol as entrainer was the best option in terms of total annual costs, and a practical implementation using a dividing wall is proposed.

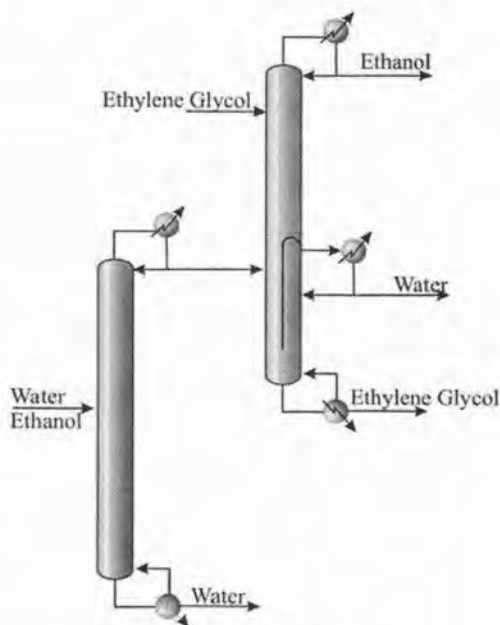


Figure 15. Practical implementation suggested for the ETCDS-SR.

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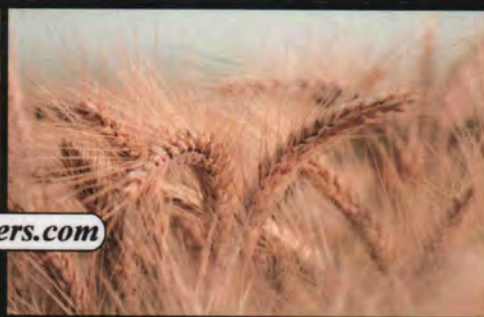
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