Comparative Study of Energy-Integrated Distillation Systems Based on Exergy Analysis and Greenhouse Gas Emissions

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The distillation is a key separation technology for the chemical industry. Its disadvantage is the large energy consumption which involves high costs and emissions associated with the energy production. Three different energy-integrated distillation schemes are investigated: the direct sequence with backward heat-integration (DQB), fully thermally coupled distillation column (FTCDC), and sloppy distillation system with forward heat-integration (SQF). The studied distillation systems are compared to each other and to the non-integrated conventional arrangement in the case of the separation of ternary mixtures. The objective of this study is to compare the selected distillation structures based on the parameters calculated in the exergy analysis and environmental emission estimation. The energy requirement of the distillation is produced on fossil basis and the greenhouse gas emission is estimated as carbon dioxide equivalent (CO_2e). The comprehensive investigation demonstrates that the heat-integrated structures, like DQB and SQF, have the lowest exergy loss, the highest energy efficiency, but the DQB shows better performance on a wider range of operation. Environmental emission can be assumed to be proportional to the energy consumption of design alternatives. The average relative emission saving is the highest in the case of the heat-integrated DQB scheme and this sequence shows the most flexible range of use.

Keywords: energy-integrated distillation; exergy analysis, CO_2 equivalent emission, ternary separation

The demand for energy has been continuously increasing for years and operation units with large energy demand have become more difficult to be supplied. Reconsideration and rationalization of industrial plants is recommended. The large energy requirement of distillation can be reduced by using energy integration like heat integration, thermocoupling or heat pumping.

Energy integration is an important method for conserving energy, demonstrated by different authors[1-7]. Annakou and Mizsey [1] have studied heat-integrated schemes and fully thermally coupled distillation columns (FTCDC) by rigorous modelling and compared them to conventional schemes. They have found that FTCDC can be competitive with the heat-integrated schemes only in those cases when the concentration of the middle component is high and the A/B split is harder than B/C split otherwise FTCDC is less economic than the heat-integrated scheme.

Internally heat integrated distillation column was studied by Olujic et al [4] and compared with the vapour recompression system in the case of propylene-propane split. Their paper shows that the internally heat integrated distillation column is feasible and could be competitive to a vapour recompression system. The reduction of the total annual cost could be up to 20%.

Regarding to the energy demand of the thermally coupled distillation columns Hernandez et al. [6] have studied the Petlyuk column and six alternative arrangements for ternary hydrocarbon mixture separation. They have found that the alternative distillation systems have very similar values of energy demands and thermodynamic efficiencies.

The large energy demand of the distillation urges to study the process and to identify the energy losses. Certainly the low thermodynamic efficiency of distillation is caused not by the separation process but mainly by the work necessary to introduce and remove the heat from the column [8]. In order to investigate the energy wastes a useful tool is the exergy analysis which gives an overall view about the location and scale of energy losses in the process. Some published works in literature focus on development of exergy analysis methods and expressive parameters in order to quantify and represent the lost energy and the thermodynamic efficiency of the distillation columns [9-13]. Other research works similarly to this one, apply the basic equations of the second law of thermodynamics and calculate the thermodynamic efficiency for different distillation systems [14, 15].

Suphanit et al. use successfully the exergy loss profile combined with the real column *T-H* profile in order to locate the heat transfer across the wall of the dividing-wall distillation column. The exergy analysis can be applied also for the study of complex distillation plants to identify the process sections with the highest exergy losses and to locate process sections with exergy improvement potential [15, 16].

The use of energy efficient distillation is beneficial also from the point of view of greenhouse gas emission since it is limited in most of the countries. The CO_2 emission of distillation systems is investigated [17, 18]. The authors quantify the CO_2 emission for a propylene-propane splitting process and propose a model for the estimation of CO_2. The model calculates the emissions flow rates from furnaces, boilers and integrated gas turbines. According to the authors the CO_2 emission can be decreased by 83%, using internally heat integrated distillation column compared to conventional alternatives in the case of a state of art propylene-propane split. In our paper the CO_2 emission calculations are verified using the proposed model [18].

Our study deals with the energy-integrated distillation systems. The studied distillation schemes are:

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(i) direct sequence with backward heat-integration (DQB); (ii) fully thermally coupled distillation column, also known as Petlyuk column (FTCDC); (iii) sloppy distillation system with forward heat integration (SQF); (iv) conventional direct separation sequence without energy integration which is the base case of the comparison.

The goal of this study is to compare and rank the studied distillation structures based on the parameters calculated in the exergy analysis and environmental emission estimation. The exergy analysis investigates the energy efficiency of the distillation systems.

Environmental emission can be assumed to be proportional to the energy consumption of the design alternatives. The energy requirements of the different distillation systems are calculated. The greenhouse gas emission as CO_2 equivalent (CO_2e) is estimated assuming natural gas as heat source. Natural gas is chosen as heat source for this study because it is one of the cleanest fossil fuel based on environmental considerations.

Experimental part

Systems studied

The selected three types of energy-integrated distillation systems are studied and compared to the conventional scheme in the case of ternary mixture separation. The base case is the direct sequence without energy integration. This selection is in accordance with several authors like [2, 19]. The studied distillation systems are selected from different types of conventional and energy integrated distillation groups and the best connection is chosen from this groups referring to the column coupling and to the direction of energy integration. First distillation system is a direct sequence with backward heat integration instead of forward because of its better economic and control features [2]. The same reason is behind the choice of the forward heat integration in case of sloppy distillation structures.

Description of the distillation systems studied

Direct sequence with backward heat integration (DQB)

The basis idea of this distillation system is to use the overhead vapour from the second column of increased pressure to provide heat to the first low, usually atmospheric pressure column. The reboiler of the first column is combined with the condenser of the second (fig. 1).

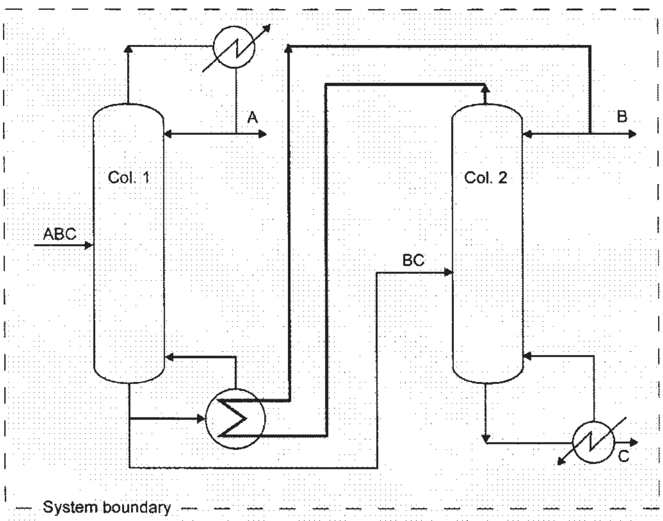

Fig. 1. Direct distillation sequence with backward heat integration (DQB)

Fully thermally coupled distillation column (FTCDC)

Fully thermally coupled distillation column also called Petlyuk column consists of a prefractionator and a main column. The required heat amount for the separation is provided through direct contact of the material flows (fig. 2). In the prefractionator it is required that the most volatile component (*A*) of the ternary mixture to be only in the top product and the heaviest component (*C*) to be only in bottom product. The middle component (*B*) distributes between the top and bottom products. The top product (V_{12} - L_{21}) and the bottom product (L_{12} - V_{21}) can be estimated [1].

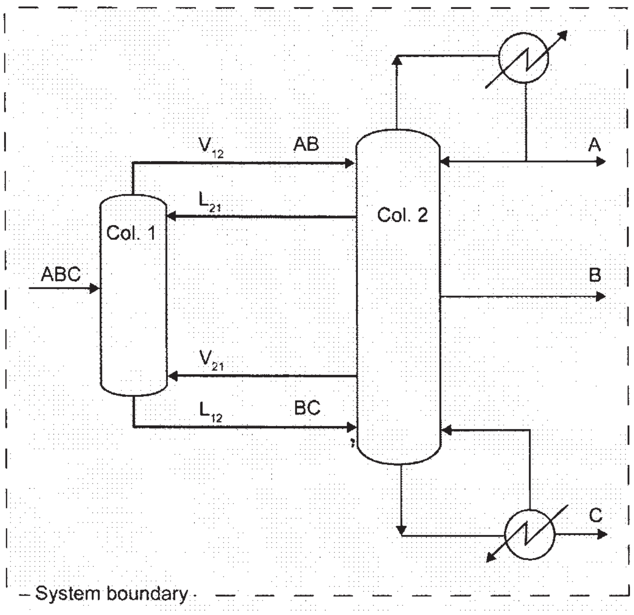

Fig. 2. Fully thermally coupled distillation column (FTCDC)

The top product of the prefractionator can be calculated with equation (1):

$$V_{12} - L_{21} = A + \beta B \quad (1)$$

The bottom product of the prefractionator can be calculated with equation (2):

$$L_{12} - V_{21} = C + (1-\beta) B \quad (2)$$

The optimal fractional recovery of the middle component estimates the flow rates in the prefractionator where the energy consumption of the FTCDC is minimal:

$$\beta = \frac{\alpha_B - \alpha_C}{\alpha_A - \alpha_C} \quad (3)$$

where β is the optimal fractional recovery of the middle component [20]; α_i is the volatility of component *i*.

Sloppy distillation system with forward heat integration (SQF)

This distillation system basically is a heat-integrated sequence, but in the prefractionator sloppy separation takes place. The middle component distribution in the prefractionator is similar to the FTCDC structure. There is no material flow from the main column to the prefractionator. The first column is a high pressure prefractionator, its distillate and bottom product are fed to the second, low-pressure column. The forward heat integration between the two columns is carried out by an integrated heat exchanger, where the overhead vapour from the high-pressure prefractionator is used to boil up the low-pressure column (fig. 3). The forward scheme is

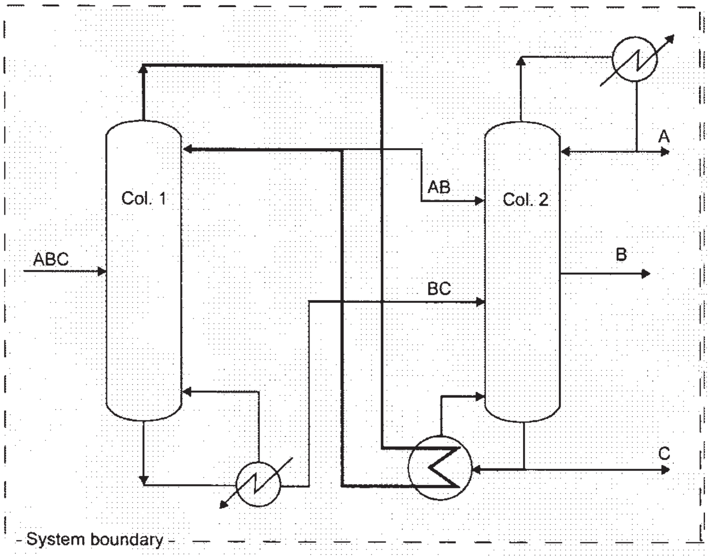

Fig. 3. Sloppy distillation system with forward heat-integration (SQF)

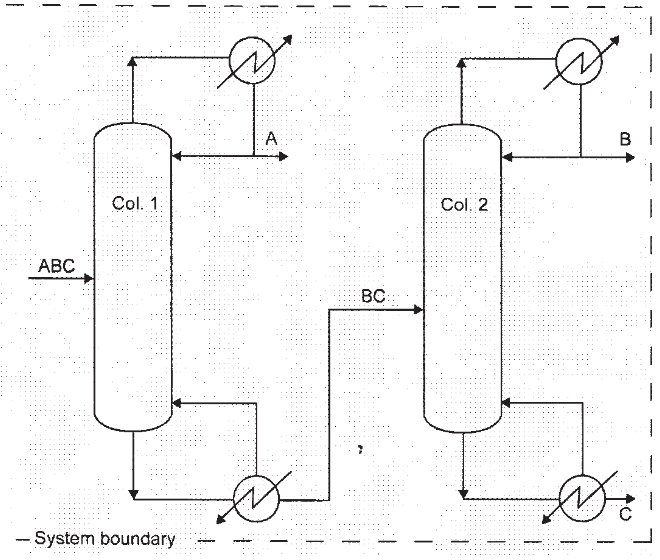

Fig. 4. Conventional direct distillation scheme (Conv. Dir.)

selected in this work because previous studies [19] have shown that it is more controllable than the backward integration.

Conventional direct distillation scheme

This separation structure is used as base case for comparison. Since it proves to be better than the indirect one in the case studies selected it consists of two simple distillation columns connected in such a way that bottom product of the first column is the feed of the second column. In literature, they are considered to be conventional arrangements for ternary distillation. Direct separation sequence is shown in figure 4.

Simulation study

Optimal parameters of the above mentioned separation systems are determined. Adequate reflux ratios, number of trays and optimal feed tray are calculated by rigorous tools. In the case studies selected (point 3), the total number of the theoretical trays of the separation schemes ranges between 70-95 and the column diameters range between 0.9-1.5 m. The reflux ratios vary according to the difficulty of separation in a wide range. The simulation models of the studied distillation systems are implemented in the ASPEN Plus process simulator. Moreover, the applied studies in this work e.g. exergy analysis requires the use of MS-Excel connected to the process simulator where the different exergy calculating equations are introduced.

Case studies

For the investigation of the distillation systems, three different ternary mixtures are selected. Three product purities are supposed (table 1-2). In the case of the exergy analysis one product purity (95%) is selected.Feed flow rate is 100 kmol/h and its composition is always equimolar. The chosen ternary mixtures have different ease of separation that can be characterized by the separation index (SI):

$$SI = \frac{\alpha_{AB}}{\alpha_{BC}} \qquad (4)$$

where α_{ij} is the relative volatility.

In mixture 1 *A*/*B* separation is more difficult than *B*/*C* separation (SI<1). In mixture 2 *A*/*B* separation is as difficult as *B*/*C* separation (SI~1), the ease of separation is balanced. In mixture 3 the *B*/*C* separation is more difficult than *A*/*B* separation (SI>1). The Soave-Redlich-Kwong model is used to calculate vapour-liquid equilibrium. This model is widely used in the refinery and gas processing industries for the prediction of vapour-liquid equilibria for systems containing non-polar components.

Exergy analysis

Energy efficiency of the studied distillation systems can be calculated based on the first law of thermodynamics which conducts to an energy analysis. Since not all the heat energy can be converted to useful work, stated by the second law of thermodynamics, exergy analysis proves to be more adequate to determine the thermodynamic efficiency of the distillation systems. By definition, exergy is the maximum capacity of the system to perform useful work as it proceeds to a specific final state in equilibrium with its surroundings.

In the case of distillation systems the columns can be seen as heat engines and their exergy demand is the minimum isothermal work of separation. Instead of mechanical work, separation work is produced.

Table 1
TERNARY MIXTURES STUDIED

Case	Mixture	α_A	α_B	α_{AB}	β	SI
1	isopentane-pentane-hexane	3.62	2.78	1.3	0.68	0.47
2	pentane-hexane-heptane	7.38	2.67	2.76	0.26	1.03
3	butane-isopentane-pentane	2.95	1.3	2.26	0.154	1.74

Table 2
EXPECTED PRODUCT PURITIES

Case	Product Purities (%)
1	99
2	95
3	90

The purpose of this analysis to compare the energy saving performance of the investigated distillation systems based on two indicators: i.) exergy loss and ii.) thermodynamic efficiency. The former gives information about the energy wastes and the latter shows the efficiency of the supplied heat conversion into separation work in the distillation systems.

The distillation process has good potential for improvement regarding energy saving when it has considerable exergy losses and low thermodynamic efficiencies [13].

In order to perform the exergy analysis the balance regions has to be fixed. Figure 1-4 shows the system boundaries where the exergy analysis is applied. The analysis takes into account the inlet and outlet material streams and the heat duty of the reboilers and of the condensers. In order to compare the thermodynamic efficiency of the studied distillation systems the entering and leaving stream physical properties must be identical. In this study the temperature, pressure, and composition of the inlet and outlet streams are equal for each case study and the product purity of the outlet streams is selected to be 95%. Therefore the inlet and outlet stream exergies are identical for the studied distillation systems separating one specific ternary mixture. Thus the separation work does not vary in function of the used distillation system.

The thermodynamic efficiency can be calculated [21] with the following equation :

$$\eta = \frac{W_{SEP}}{Ex_{loss} + W_{SEP}} \qquad (5)$$

where W_{SEP} [kW] is the work of separation, ExC_{loss} [kW] is the exergy loss of the system.

The separation work can be defined with the equation (6).

$$W_{SEP} = \sum_{outlet} nEx - \sum_{inlet} nEx \qquad (6)$$

where n [kmol/h] is the mole flow of the inlet and outlet streams, the Ex [kJ/kmol] is the specific exergy which can be calculated with the equation (7).

$$Ex = H - T_0 S \qquad (7)$$

where H [kJ/kmol] is the molar enthalpy, S [kJ/kmol K] is the molar entropy and T_0 [K] is the ambient temperature which is fixed at 283 K in this work.

Gouy-Stodola theorem states that the lost available work is directly proportional to the entropy production. The proportionality factor is simply the ambient temperature T_0:

$$Ex_{loss} = T_0 \Delta S_{irr} \qquad (8)$$

Based on the second law of thermodynamics the entropy production can be calculated:

$$\Delta S_{irr} = \sum_{outlet} (nS + \frac{Q_{cond}}{T_{cond}}) + \sum_{inlet} (nS + \frac{Q_{reb}}{T_{reb}}) \qquad (9)$$

where Q_{cond} and Q_{reb} [kW] are the heat duties of the condenser and reboiler, T_{cond} and T_{reb} [K] are the temperatures of the cooling and heating media, respectively. The cooling water temperature is taken 283 K and the temperature of the steam used in the reboilers is 433 K.

The calculated thermodynamic efficiency is linear dependent on the taken temperature grades [22] thus one can obtain different efficiency values if the temperature of the used cooling and heating media is different or the ambient temperature is fixed differently.

Results of the exergy analysis

Exergy analysis is applied to the studied energy-integrated distillation systems and to the conventional direct distillation scheme. Each distillation system is studied for three different ternary mixtures (table 1.) in order to investigate also the effect of SI on the energy saving properties of the distillation systems. The distillation systems are ranked according to their exergy loss and thermodynamic efficiency.

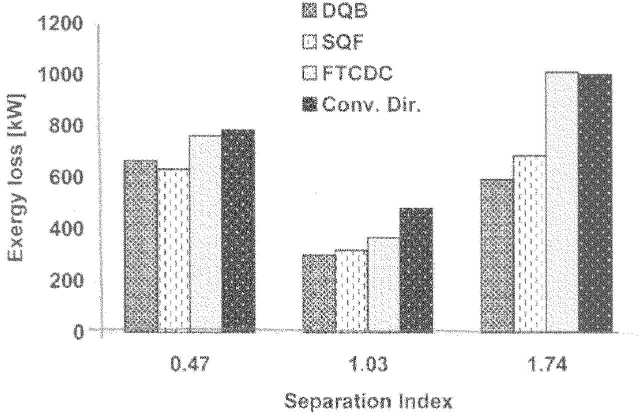

Fig. 5. Exergy loss function of the separation index

In figure 5 the exergy loss is presented in function of SI for each distillation system. The three SI value represents the three ternary mixtures with different ease of separation. As the figure shows the exergy loss is the lowest when the ease of separation of the mixture is balanced so the SI value is close to 1. If the ease of separation becomes unbalanced thus the SI of the mixture differs from 1 the exergy loss increases. The exergy loss of the distillation systems generally is the highest in the case of the third mixture separation with SI=1.74. It has to be noted that SI value of the third mixture deviates the most from 1 which means that this mixture is the furthest from the symmetric ease of separation.

The comparison of the different distillation systems shows that the DQB has the least exergy loss followed by SQF. Entirely these two heat integrated distillation systems show the best energy saving and consequently the thermodynamic efficiency of these systems is the highest (figure 6). The thermodynamic efficiency of the FTCDC is better compared to the conventional direct scheme in the case of the mixtures with SI=1.03 and in the case of the other mixtures there is very slight difference in the thermodynamic efficiency of the FTCDC and conventional direct scheme. Figure 6 shows that the DQB has 5% higher thermodynamic efficiency than that of the conventional direct scheme in the case of mixture 2 with SI=1.03. These results support the idea that processes with high exergy loss and low thermodynamic efficiency can be energetically improved [13].

The results of the exergy analysis are compared to the energy consumption data of the distillation systems. The calculation of the energy consumption assumes that the distillation systems have only heat requirements neglecting e.g. the electricity usage of the pumps etc. The heat demand of the studied distillation systems is presented on the figure 7. One can notice that the tendency of the heat demand is similar to that of the exergy analysis. This can be explained with the identical ambient and utility temperatures for each case study. The exergy analysis

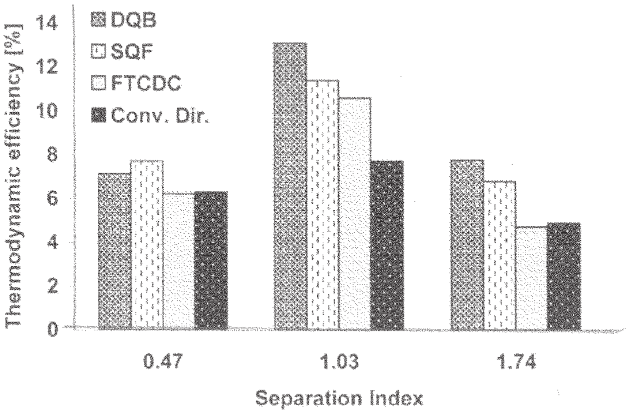

Fig. 6. Thermodynamic efficiency function of the separation index

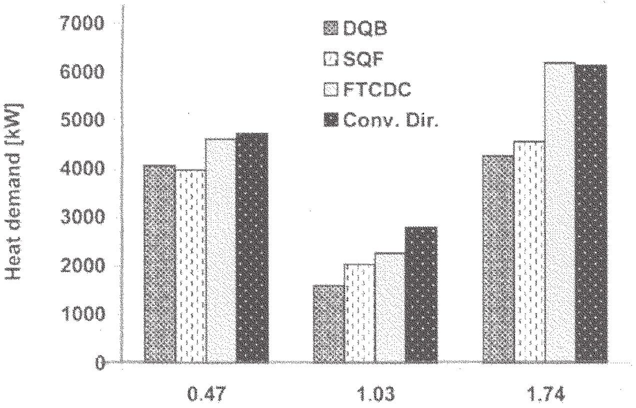

Fig. 7. Heat demand of the distillation systems in function of the separation index

Table 3
GLOBAL WARMING POTENTIALS OF DIFFERENT GREENHOUSE GASES

Greenhouse Gases	GWP value / 100 years
Nitrous oxide (N_2O)	296
Methane (CH_4)	23
Trifluoromethane (HFC-23)	12000
1,1,1,2-Tetrafluoroethane (HFC-134a)	1300
Sulfur hexafluoride (SF_6)	22200

reveals the characteristics of the studied distillation systems regarding to the energy efficiency but it is uncertain how precisely the CO_2e emission of these systems can be predicted. Therefore the next step of this work is the estimation of the CO_2e emission of the studied distillation systems.

Estimation of carbon dioxide equivalent (CO_2e) emission

Since the European Union ratified the Kyoto Protocol and established in 2005 a scheme for greenhouse gas emission trading, carbon dioxide emission is of especial importance. Some countries have introduced taxes based on the carbon content of the energy products and this tax is called 'carbon tax'. A 'carbon tax' is a charge to be paid on each fossil fuel, proportional to the quantity of carbon emitted when it is burned. Concerning the present demands, carbon dioxide equivalent emissions need to be quantified.

Reducing energy requirements of distillation systems leads to lower CO_2 emission, this is why reconsideration of distillation plants is necessary. Energy integration could be a solution to reach a lower CO_2 emission through better energy utilization. The environmental emissions of the studied energy-integrated distillation systems are estimated and compared to the emission of conventional distillation scheme. Therefore the energy requirements of the different distillation systems are calculated. Based on the energy consumption data of the distillation schemes, environmental emissions are estimated using SimaPro 6.0 and the results are verified [18] for quantifying the emissions associated with heat integrated distillation systems. Beside of CO_2 emission other greenhouse gases are also estimated like nitrous oxide (N_2O), methane (CH_4), hydrofluorocarbons, and sulfur hexafluoride (SF_6). Carbon dioxide equivalent is used in order to quantify these emitted greenhouse gases. The global warming potential (GWP) values of the greenhouse gases are taken for 100 years time horizon (table 3).

The CO_2e is calculated by summing the greenhouse gas emissions multiplied by their GWP value (10).

$$CO_2\ equivalent\ \ emission = \sum (GWP \times Greenhouse\ \ gas\ \ emission) \tag{10}$$

The CO_2e gas emissions are estimated assuming natural gas as heat source. The emitted greenhouse gases using this fossil fuel are composed mainly of CO_2 and of slight amount of N_2O and CH_4. The amount of emitted halogenous compounds is practically negligible compared to the CO_2 emission and therefore it can be neglected. Appropriate firing equipment is selected for natural gas based on the descriptions found in the literature [23]:natural gas heating atmospheric burner with low NO_x emission with 87% efficiency.

Results of CO_2e emission estimation

The investigated energy-integrated distillation systems have lower CO_2e emission compared to the conventional direct distillation scheme. The CO_2e emission reduction compared to the base case is also investigated. The CO_2e emissions are summarized for all the three product purities into one average CO_2e emission saving figure. These figures are shown in one chart (fig. 8) for all the three studied energy-integrated distillation schemes.

Figure 8 shows that the DQB structure has the best figure. This means that the DQB scheme is the most flexible in the CO_2e emission issue if product purity changes. The average emission saving of DQB reaches 40%.

The CO_2e emission confirms the results of the previous study of this work and draws the attention to the consequences of wasted energy.

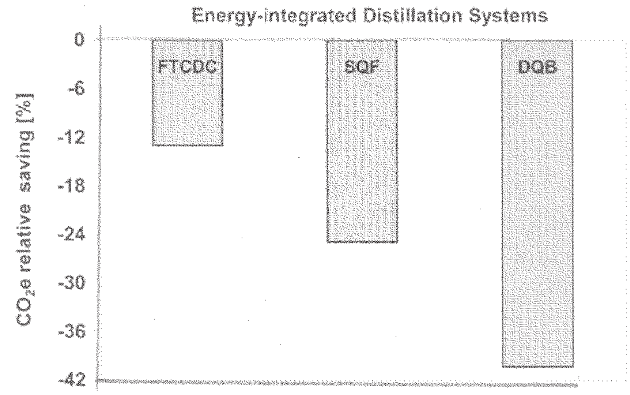

Fig. 8. CO$_2$e relative emission saving correlate to the conventional direct distillation scheme

Conclusions

The effect of energy-integration is studied in the case of distillation system analyzing their thermodynamic efficiency and CO$_2$e emission. Heat-integrated and thermally coupled distillation systems are compared to each others and to the non-integrated conventional arrangement as base case.

Based on the exergy and CO$_2$ emission study, this paper can help for choosing the adequate energy-integrated distillation variant in case of ideal hydrocarbon mixtures.

In the case of the exergy analysis the base of comparison is their exergy loss and thermodynamic efficiency. This analysis shows that the heat-integrated distillation structures (DQB and SQF) are the most energy-efficient among the studied ones. The thermodynamic efficiency of all the studied distillation systems is the highest when the ease of separation is symmetric. CO$_2$e emission reduction can be achieved using heat-integrated distillation schemes. CO$_2$e emissions of the distillation systems can be decreased by 40% using DQB arrangements instead of conventional alternatives. The CO$_2$e emission estimation demonstrated that in most of the case DQB has the lowest CO$_2$e emission but the SQF is less sensitive to the product purity change.

It can be concluded that the energy integration saves energy, and as a consequence CO$_2$e emission, too. Among the different distillation schemes the heat-integrated DQB alternative proves to be the best applicable since it shows the features in a wide and flexible range.

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Nomenclature

Latin symbols

A - The most volatile component of the ternary mixture
B - Middle component of the ternary mixture
C - The heaviest component of the ternary mixture
CO$_2$e - Carbon dioxide equivalent, [kg]
DQB - Direct distillation sequence with backward heat integration
Ex - Specific exergy, [kJ/kmol]
Ex$_{loss}$ - Lost exergy rate, [kW]
FTCDC - Fully thermally coupled distillation column
GWP - Global warming potential
H -- Molar enthalpy, [kJ/kmol]
L$_{12}$ - Liquid flow rate from column 1 to column 2, [kmol/h]
L$_{21}$ - Liquid flow rate from column 2 to column 1, [kmol/h]
n - Mole flow, [kmol/h]
Q$_{cond}$ - Heat duty of the condenser, [kW]
Q$_{reb}$ - Heat duty of the reboiler, [kW]
S - Molar entropy, [kJ/kmol K]
SI - Separation Index
SQF - Sloppy distillation system with forward heat-integration
T$_0$ - Ambient temperature, [K]
T$_{cond}$ - Cooling water temperature, [K]
T$_{reb}$ - Heating steam temperature, [K]
V$_{12}$ - Vapour flow rate column 1 to column 2, [kmol/h]
V$_{21}$ - Vapour flow rate from column 2 to column 1, [kmol/h]
W$_{SEP}$ - Work of separation, [kW]

Greek letters

α_i - Volatility of component i
α_{ij} - Relative volatility of components i, j
β - Optimal fractional recovery of the middle component
ΔS_{irr} - Entropy production [kJ/h K]
η - Thermodynamic efficiency

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